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UNITED STATES DEPARTMENT OF ENERGY
UNDER CONTRACT NUMBER DE-AC06-76RL01857

RADIONUCLIDE INVENTORY AND SOURCE TERMS FOR THE SURPLUS PRODUCTION REACTORS AT HANFORD

Edited by R. L. Miller and J. M. Steffes

DISTRIBUTION

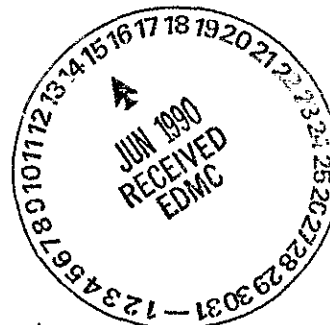| NAME	BLDG/RM/MS	NAME	BLDG/RM/MS
DE-RL		20. BJ Carlson	1720K/K3
RE Austin	FED/640	21. JJ Dorian	1114N/21/S3
JD Goodenough	FED/303	22. KA Gano	1114N/16/S3
JR Hunter	FED/607B	23. PW Griffin	1720K/2A/K3
CE Miller, Jr.	FED/302A	24. LL Grumme	1104N/29/E7
PE Rasmussen	FED/600	25. MC Hughes	1721K/K9
JH Slaughter	FED/506	26. BW Mathis	1720K/K1
IC		27. RL Miller	FED/379/F21
HF Daughtery	FED/472	28. EW Powers	1720K/K3
MW Young	337/3/S1	29. DL Renberger	1117N/13/C1
IL		30. SE Robyler	1117N/51/C12
EB Moore, Jr.	Sigma 2/71	31. DR Skeen	1117N/51/C12
ES Murphy	209E/16	32. GL Smith	1118N/22/B2
BA Napier	2400 Stevens	33. JM Steffes	1721K/K9
WL Templeton	Life Sci/331	34. RA Winship	FED/376/F21
ckwell		35. UNI File	FED/436/F11
WF Heine	2750E/D101	36. UNI Record	FED/436/F11
JH Roecker	2750E/B210	37. Extra	
DR Speer	1720K/K3	38. Extra	
IC		39. Extra	
JA Adams	FED/300H/F27	40. Extra	
JF Beckstrom	1720K/Lab/K3	41. Extra	
DF Brendel	1101N/121/N2	42. Extra	
HM Burton	FED/466/F19	43. Extra	

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SUMMARY

Radionuclide inventories have been estimated for the eight surplus production reactors at Hanford. The inventories listed represent more than 95% of the total curie burden; the remaining 5% is distributed in piping, tunnels, and various other locations within the reactor building and unaccounted for inventories within the reactors or fuel storage basins. Estimates are conservative as the methodology was designed to overestimate the radionuclide inventories in the facilities.

The estimated inventory per reactor facility ranges from 13,000 curies to 58,000 curies. The majority of the present inventory consists of tritium, carbon-14, cobalt-60, and nickel-63.

The information in this document combines data from past characterization efforts and introduces adjustments for added information and refinement.

Since the reactors have been shut down from 15 to 20 years, many of the shorter half-life radionuclides have decayed to insignificant levels and are therefore not addressed. Trace amounts of some longer half-life radionuclides are listed only to show that they were evaluated.

The inventory of hazardous materials in the reactor facilities is also addressed.

The document has been revised to include new reduced inventory figures for chlorine-36. The new figures were derived from recent analysis of irradiated graphite from the 105-KW reactor.

ACKNOWLEDGEMENTS

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1.0 INTRODUCTION

1.1 PURPOSE

The purpose of this document is to provide estimated inventories of radionuclides and other hazardous materials in the eight Hanford 100 Area surplus production reactor buildings. This information is intended to support the preparation of an Environmental Impact Statement (EIS) currently being prepared by Battelle Pacific Northwest Laboratory (PNL) for the final decommissioning of these facilities.

1.2 SCOPE

The estimated reactor radionuclide inventories are based on previous analysis and physical measurements. Supporting documentation is included in the appendices and references. Hazardous material inventories are based upon recent walk-through inspections of each facility.

For this document, each reactor facility was grouped into three sections: 1) the reactor block, which includes the graphite moderator stack, biological and thermal shields, process tubes, and the safety and control systems; 2) the irradiated fuel storage basin; and 3) contaminated portions of the facility that are outside the reactor block and fuel storage basin but are within the reactor building decommissioning boundary.

Where numerical data contained in this report were estimated, the estimates are conservative. They were designed to overestimate the radionuclide and hazardous material inventories present. Impacts of special tests, special fuel loadings, special control rods, etc., were not calculated in the inventory estimates since their effect on the overall inventory is not considered significant. For the same reason, no attempt was made to examine the spatial effects that occur due to fuel, control rods, test holes, special loadings, etc. Prior to actual decommissioning a detailed characterization of

each reactor facility will be performed as necessary to provide definitive estimates of waste volumes, radiation dose rates, and radionuclide inventories.

At the time the reactor decommissioning planning documents were prepared, the date of March 1, 1985, was selected as a target for beginning decommissioning work. The date has been maintained in this document for decay corrections to inventory figures.

All radionuclide inventory values are expressed in curies of the particular isotopes present. Readers are advised that even though values for some radionuclides are in the thousands of curies while others are extremely small, in some instances the small value is more important environmentally than the large value, due to such factors as half-life period and biological effects. This is especially true for transuranic radionuclides, which have very large curie to rem (roentgen-equivalent-man) conversion factors.

1.3 HISTORY OF HANFORD 100 AREA REACTORS

The Hanford Site (Figure 1) occupies 570 square miles in southeastern Washington State and was commissioned in 1942 for the production of plutonium by the Manhattan Engineering District of the U.S. Army Corps of Engineers. Eight graphite-moderated reactors and associated support facilities were constructed in the Hanford 100 Area between 1942 and 1954 to support the national defense plutonium production effort. The reactors were designated B, C, D, DR, F, H, KE, and KW. The reactors were shut down between 1964 and 1971. Table 1 summarizes the reactors' operating histories.

A ninth production reactor, N Reactor, was started up in 1963 and is still in operation. Not considered for decommissioning at this time, N Reactor is not included in this report.

During the operating life of the reactors, several changes were made in the hardware and fuel loadings to support operation at higher power levels and greater production efficiency. These changes have significantly influenced
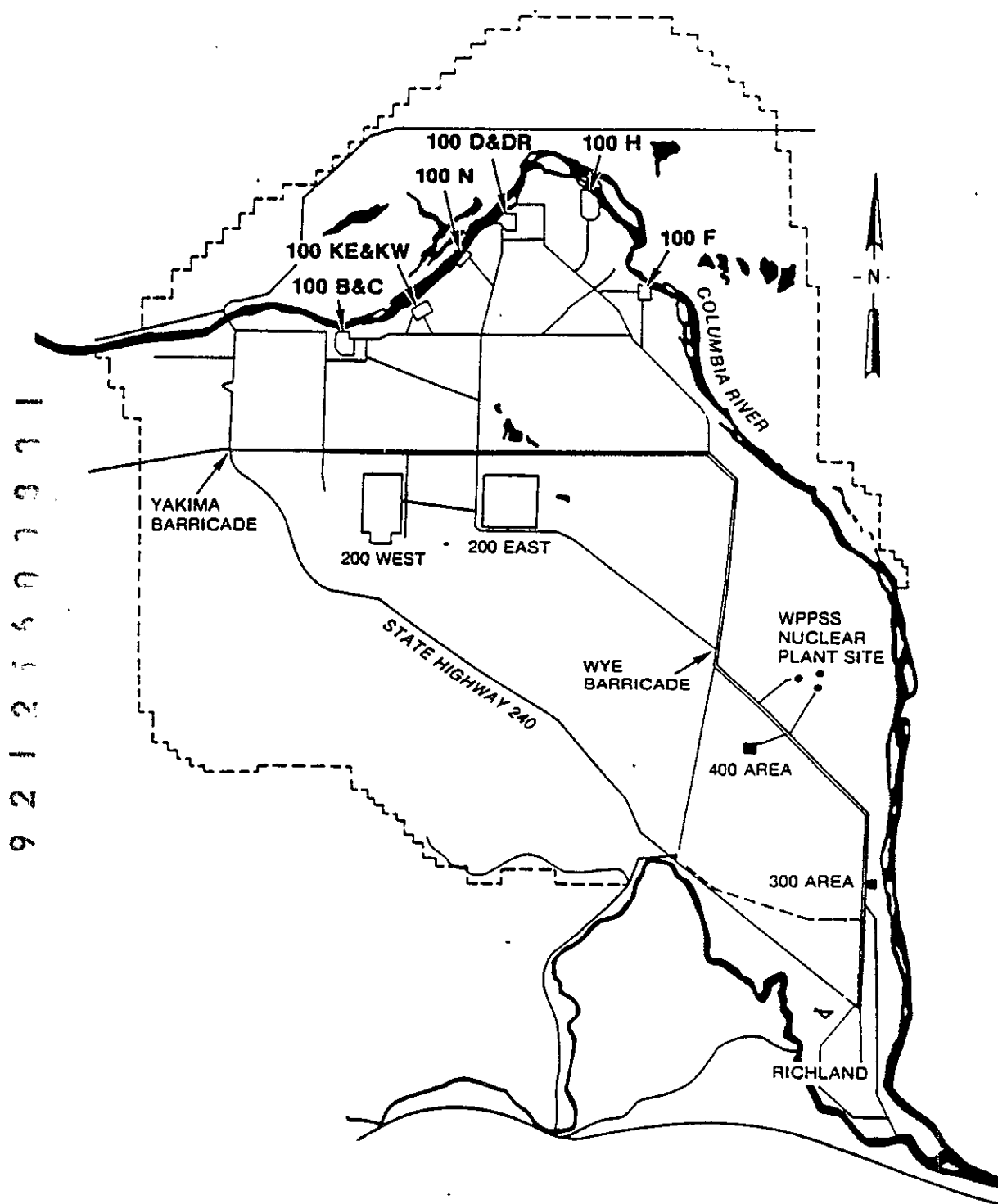
Figure 1. Hanford Site. The eight production reactors are situated along the Columbia River.

TABLE 1

REACTOR OPERATING HISTORIES

Reactor Facility	Initial Startup Date	Final Shutdown Date	Years Operated
B	09/26/44	02/13/68	22*
C	11/18/52	04/25/69	17
D	12/17/44	06/26/67	23
DR	10/03/50	12/30/64	14
F	02/25/45	06/25/65	20
H	10/29/49	04/21/65	16
KE	04/17/55	01/28/71	16
KW	01/04/55	02/01/70	15

*B Reactor was shut down and held in standby status from 03/19/46 to 06/02/48, then restarted and operated until February 1968.

the quantities and distribution of radionuclides remaining in the reactors. Higher equilibrium power levels in the older reactors were achieved during the early 1950's by using various control rod patterns and installing neutron-absorbing material in some of the process tubes in place of fuel. At the same time, some enriched fuel loadings (higher uranium-235 concentration) were introduced to increase the flattening or evening-out of the power levels. Then in the late 1950's even more dramatic power level increases were achieved in the six older reactors by introducing an "enrichment ring" and significantly increasing the cooling water flow rate through the reactors. The enrichment ring consisted of several hundred enriched fuel columns installed in a ring pattern near the outside of the process tube pattern. The effect was to create a central zone of the reactor that had nearly equal individual tube powers. Prior to loading the enrichment rings, the power

level of the reactor was determined by the "hottest" tubes, which were usually located near the center of the reactor. With the flattened power distribution, the limiting condition for the older reactors became the bulk outlet coolant temperature for the entire reactor.

The overall effect of flattening the power in the central zone of the reactor and increasing the cooling water flow resulted in final equilibrium power levels in the 2000 MW range for the older reactors, which had design power levels of 250-350 MW, and a final level of 4400 MW for the K reactors, which had design power levels of 2500 MW.

1.4 DOCUMENT ORGANIZATION

The major portion of the document is divided into three sections corresponding to the groupings of a reactor facility: the reactor block, the irradiated fuel storage basin, and the other areas of the building that might contain some radionuclides. Within each of the three sections, the methods and sources of characterization data, the calculation adjustments and assumptions, and the inventory estimates are presented.

The information in this document combines data from past characterization efforts and introduces adjustments for added information and refinement.

In order to determine a conservative and realistic radionuclide inventory estimate, several general assumptions were made to simplify the complexity of the materials and conditions in the reactors. These simplifying assumptions and the adjustments, along with a rationale, are provided in the section under the reactor divisions. In all cases, conservative assumptions and adjustments were applied.

The radionuclide inventories were estimated for the major components of the surplus reactors. An attempt was made to provide two significant figures for each estimate. In some cases the uncertainty involved in making the estimate

precluded the use of the two significant figures. An example of the uncertainty is the calculation of trace nuclides in the shields where the parent nuclide concentration was estimated from handbook values for "typical" materials.

Revision 1 of this document was made to include reduced chlorine-36 estimates derived from measurements of actual irradiated graphite samples as opposed to the original estimates based on unirradiated graphite samples.

2.0 REACTOR BLOCK

The reactor block (Figure 2) is located near the center of the reactor building. Horizontal control rod penetrations are on the left side of the block (when facing the reactor front face), experimental test penetrations are on the right side. Fuel discharge and storage areas are located directly behind the rear of the reactor. The vertical safety rod and ball 3X system penetrations are on the top of the reactor. The ball 3X system is an emergency shutdown system consisting of neutron-absorbing balls that could be released into the reactor core from hoppers above the reactor.

A typical older reactor block consists of a graphite moderator stack encased in a cast iron thermal shield and a biological shield, consisting of alternating layers of Masonite and steel (Figures 3 and 4). In both of the reactors, heavy-aggregate concrete was used as a biological shield. The reactor block rests on a massive concrete foundation.

The principal components of a production reactor block are:

o The reactor moderator stack, which is an assembly of graphite blocks cored to provide channels for process tubes, control rods, and other equipment.

o Process tubes, which contained the uranium fuel elements and provided channels for cooling water flow.

o Horizontal control rods.

o Vertical safety rods.

o Ball 3X system, for dropping neutron-absorbing balls into vertical safety rod channels for emergency reactor shutdown.

o Monitoring and experimental test equipment.

o Thermal and biological shielding, surrounded by a heavy, vault-like steel outer shell equipped with gas-tight seals for the reactor block penetrations.
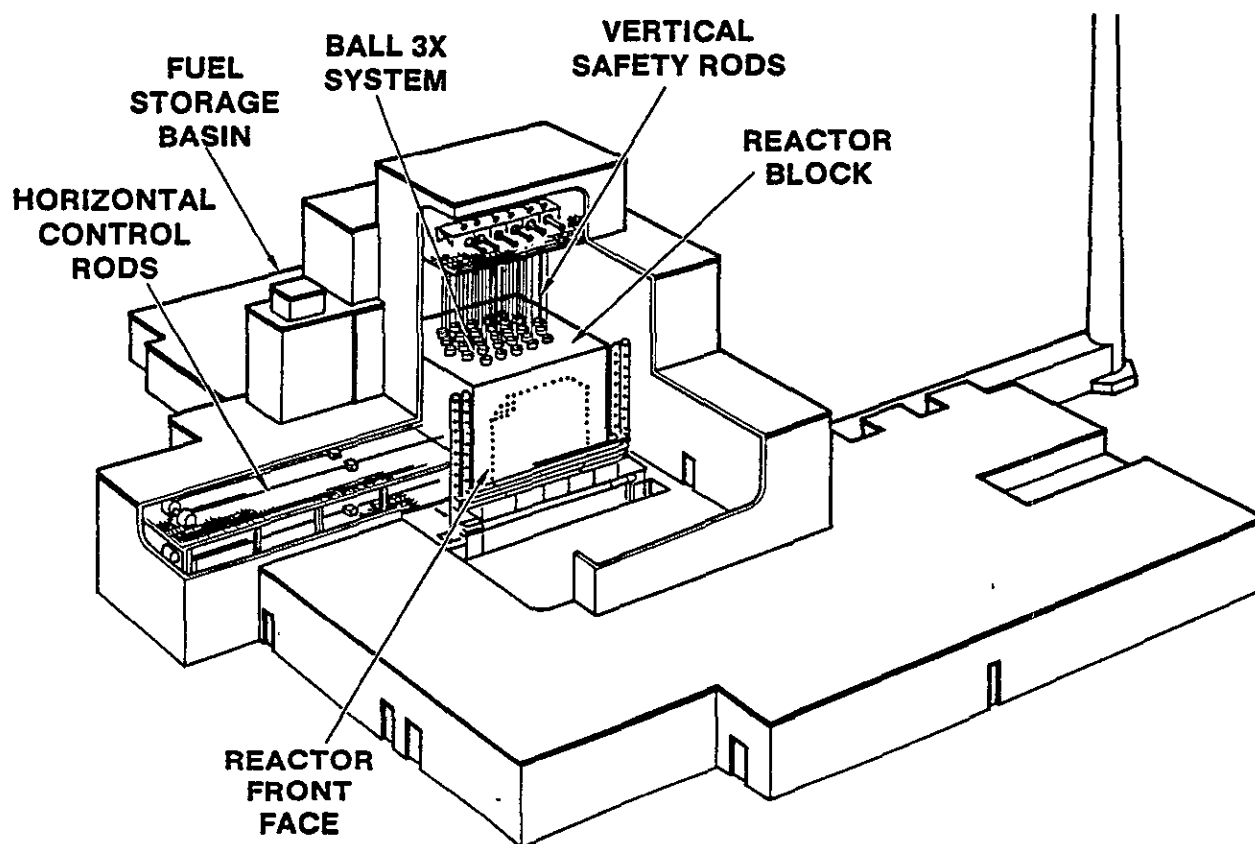
Figure 2. Typical Reactor Facility.

For inventory purposes, the reactors are assigned to two groups: 1) the older reactors B, C, D, DR, F, and H; and 2) the newer KE and KW reactors. The older reactors' thermal shields and graphite stacks are of similar size and were constructed of similar materials. The older reactors also had similar neutron flux distributions.

The two K reactors are comparable to the older reactors, having been constructed from similar drawings and specifications. But they had graphite stacks 1-1/2 times larger than the older reactors, larger fuel elements, more

REACTOR BLOCK
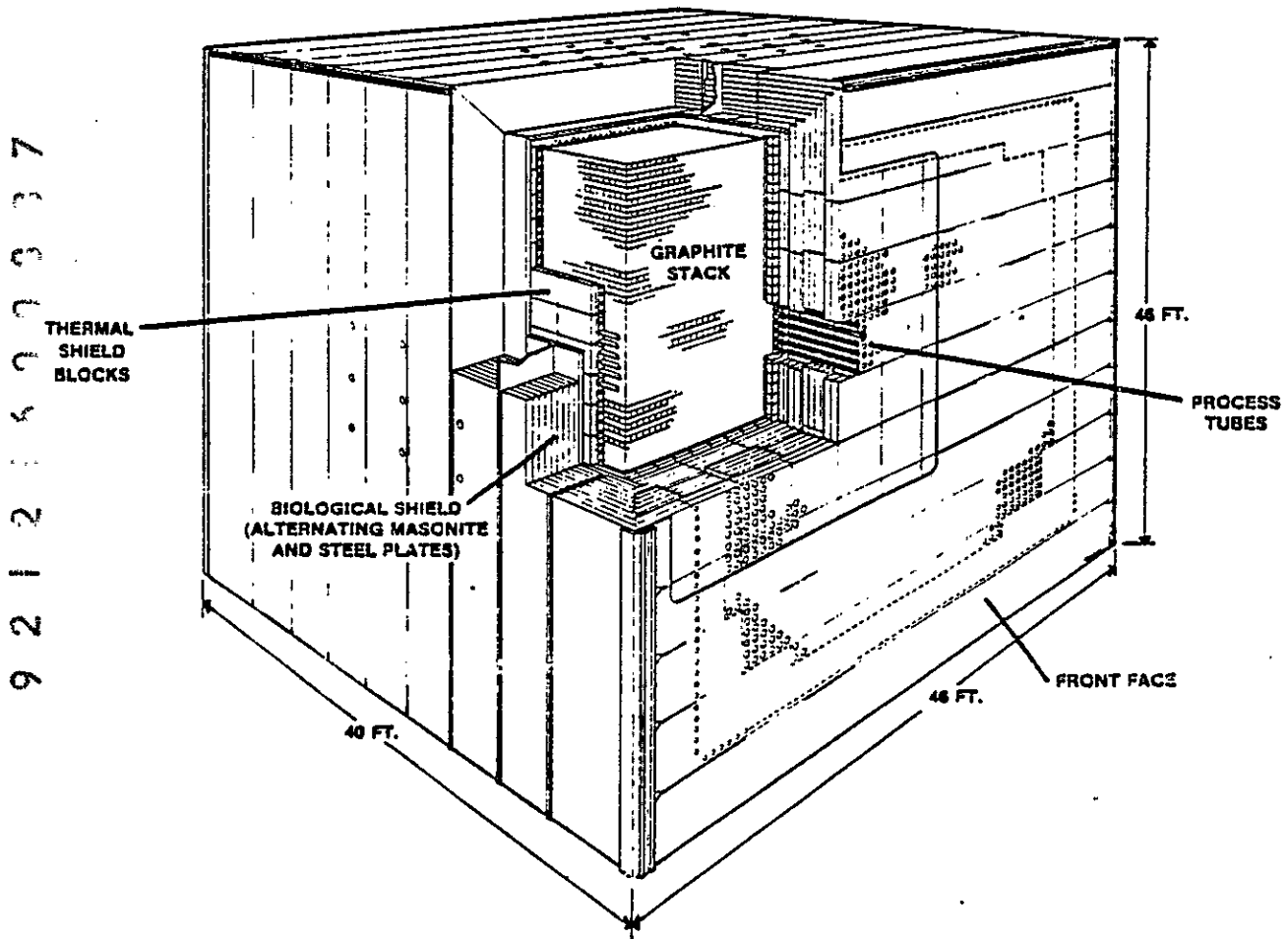
Figure 3. Reactor Block Construction.
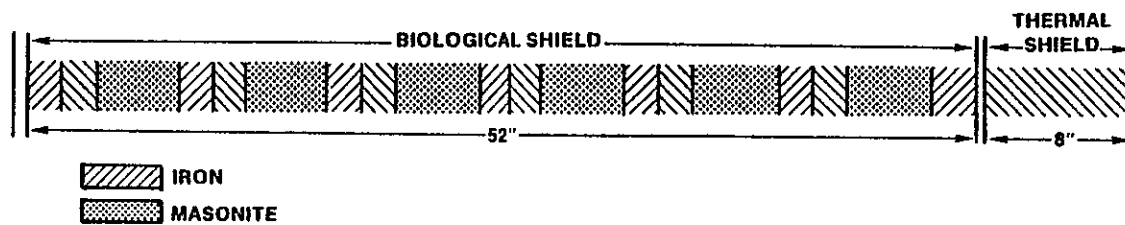
Figure 4. Cross Section of Biological and Thermal Shields.

and different-material process tubes, and heavy-aggregate concrete biological shields. Additionally, the two K reactors were operated at a higher power level. Table 2 shows the comparison of the reactor groups.

TABLE 2

HANFORD PRODUCTION REACTOR DESIGN DATA

| Reactors | Graphite Stack Dimensions (ft) | | | Process Tubes | | | Thermal Shield | | Biological Shield | |
	Front to Rear	Top to Bottom	Side to Side	Number	Type	ID (in.)	Type	Thickness (in.)	Type	Thickness (in.)
B, C,* D, DR, F, H	28	36	36	2004	Aluminum	1.75	Cast Iron	8-10	Steel and Masonite	52
KE, KW	33.5	41	41	3220	Zircaloy and Aluminum	1.8	Cast Iron	10	Heavy-Aggregate Concrete	45-83

*C Reactor has slightly larger diameter process tubes than the other reactors in this group. It has a heavy-aggregate concrete top biological shield (84 in. thick) in place of steel and Masonite.

2.1 METHODS AND SOURCES FOR REACTOR BLOCKS

The main source of characterization data for the reactors is recorded in the DR Reactor Characterization (Appendix A). The radionuclide concentration data in Appendix A are used in this document to derive estimated inventories of radionuclides for the DR Reactor. Inventory numbers for the other older reactors (B, C, D, F, and H) are based on the characterization results from the core sampling of the DR Reactor block, then adjusted for operating history and fluence differences. Inventory numbers for the K Reactors were based both on some sample analyses and by adjusting the DR Reactor data.

Where sample analysis data were not available or did not appear adequate for the specific component, a calculation was made using basic activation computational techniques or using the data set presented in Reference 1.

2.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR REACTOR BLOCK

The principal components of each reactor block are the graphite moderator stack, the cast iron thermal shields, the process tubes (which contained the fuel charges); the control rods, safety rods, emergency shutdown ball 3X system, and the biological shields. Each of these components is addressed separately in paragraphs 2.2.1 through 2.2.5.

2.2.1 Graphite Moderator Stack

The graphite moderator stack consists of 4-foot-long graphite blocks stacked to provide a central region for fuel loading and an outer region for a neutron reflector. Each of the older reactors (B, C, D, DR, F, and H) contains 2004 process tube openings; the two K Reactors each have 3220 process tube openings. In addition to these openings, the graphite block has openings for control rods, safety rods, test facilities, and instrumentation. The original four Hanford reactors (B, D, DR, and F) are very nearly identical; the DR Reactor has an additional test facility. The C and H Reactors have more control rods (15 versus 9), more safety rods (45 versus 29) and more test facilities (13 at C Reactor, 10 at H Reactor, 3 each at B, D, F, and 4 at DR). Each K Reactor has 20 control rods, 45 safety rods, and 16 test facility openings.

As discussed in Section 1.3, at the end of their operating life, each reactor had an enrichment ring installed to provide power flattening in the central zone of the reactor. It should be noted that the flattening of power in the central zone affected the neutron flux levels only in the radial direction from an axis in the front-to-rear direction. The axial flux distribution was not flattened except to a very limited extent achieved by judicious placement

of control rods. The axial flux distribution was closely approximated by a cosine distribution in the front-to-rear direction. This cosine shape of the flux allowed another convenient means of applying a conservative factor, i.e., assume that the flux level is constant at the peak level over the entire fuel charge length. This factor of conservatism was used whenever a calculation was used in place of sample data.

An additional conservative adjustment was made in the radial direction. Although the enrichment ring was several lattice units (process tubes) in from the outside of the charge pattern, for calculation purposes the flattened zone was assumed to extend to the entire active region of the reactor.

Table 3 summarizes the characteristics used for the reactor graphite stacks.

Radionuclide inventories in the graphite stack were determined for the DR Reactor by using the average concentration values presented in Appendix A, decay-correcting to March 1, 1985, and multiplying by the stack mass. A similar procedure was used for the K reactors' inventories using KW sample analysis data (Appendix B).

The estimated radionuclide inventories for the old reactors, other than DR, were calculated by taking the ratio of the neutron fluences or the total effective full-power operating days as a multiplier for the DR Reactor data. The values obtained were decay-corrected to March 1, 1985. Table 4 shows the equivalent full-power days operated, the fluence, and the decay times for each reactor.

TABLE 3

REACTOR STACK PHYSICAL CHARACTERISTICS

Parameter	Old Reactors	K Reactors
Dimensions of Stack:		
Side to Side	36 ft (11 m)	41 ft (12.5 m)
Top to Bottom	36 ft (11 m)	41 ft (12.5 m)
Front to Rear	28 ft (8.5 m)	33.5 ft (10.2 m)
Fuel charge length	23.8 ft (7.2 m)	28.1 ft (8.6 m)
Number of process tubes (fuel charges)	2004	3220
Lattice spacing for process tubes (fuel charges)	8-3/8 x 8-3/8 in.	7-1/2 x 7-1/2 in.
Volumes:		
Entire stack	1028 m^3	1595 m^3
Active zone	658 m^3	1001 m^3
Reflector	370 m^3	594 m^3
Process tubes	(24 m^3)*	(45.3 m^3)*
Control and Safety rods	(4.1 m^3)*	(4.8 m^3)*
Test facilities	(0.3 m^3)*	(1.4 m^3)*
Density of graphite	1.7 g/cm^3	1.7 g/cm^3
Mass of graphite:		
Active zone	1.07×10^9 g	1.61×10^9 g
Reflector	6.3×10^8 g	1×10^9 g
Flux levels (neutrons/cm^2-s)		
Active zone	5×10^{13}/cm^2-s	1×10^{14}/cm^2-s
Reflector (center)	5×10^{12}/cm^2-s	5×10^{12}/cm^2-s
(outer edge)	1×10^{12}/cm^2-s	1×10^{12}/cm^2-s

*Volume of openings not included in entire stack volume calculation.

TABLE 4

OPERATING DATA AND DECAY TIMES FOR RADIONUCLIDE CALCULATIONS

Reactor	Final Operating Power Level (MW)	Full-Power Days Operated	Fluence,* 10^{22} per cm^2	Decay time to 3/1/85 (yrs)
B	1960	5092	2.20	17
C	2310	5108	2.21	15.8
D	2005	4933	2.13	17.7
DR	1925	3610	1.56	20.2
F	1935	4181	1.81	19.7
H	1955	3990	1.72	19.9
KE	4400	5311	5.51	14.1
KW	4400	5043	5.23	15.1

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*The peak neutron flux assumed was $1 \times 10^{14}/cm^2$-s for the K Reactors and $5 \times 10^{13}/cm^2$-s for all other reactors.

2.2.2 Thermal Shield

A cast iron thermal shield surrounds the graphite stack. Constructed of overlapping blocks, an older reactor's shield varies in thickness (8 to 10 inches) on the sides, top, bottom, front and rear. The K Reactor thermal shields are all 10 in. thick. The inventory for the thermal shield also includes the contribution from the gunbarrels, which are steel tube connections for cooling water and instrumentation. Figure 5 is a simplified cross-section of a process tube penetration through the reactor, showing the gunbarrel, process tube, thermal and biological shields, and graphite relationships.
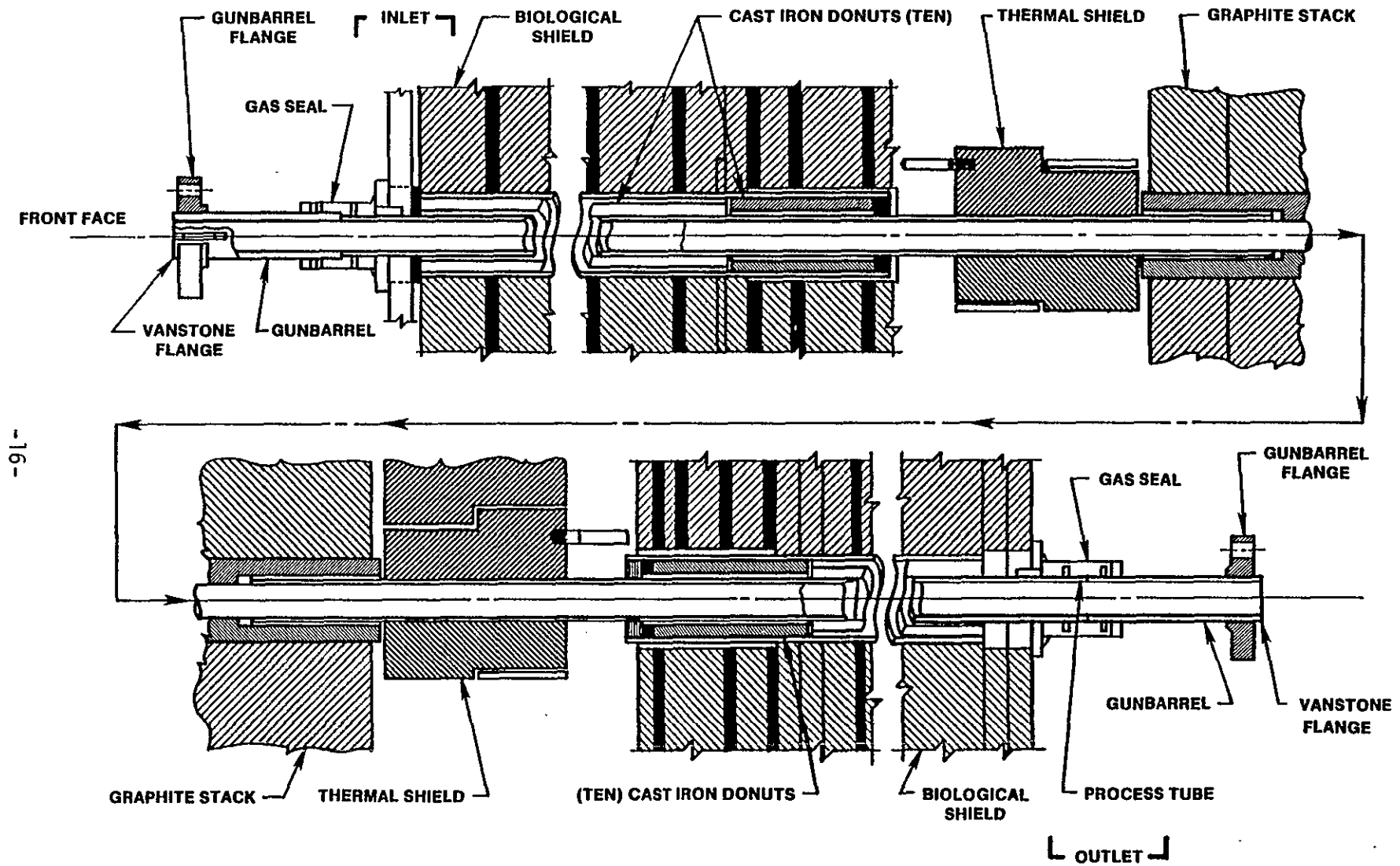
Figure 5.   Process Tube Channel Cross Section.

In addition to the radionuclide analysis of the DR Reactor thermal shield samples provided in Appendix A, a chemical analysis was performed on samples obtained from a mock-up which had been assembled using original reactor components serial-numbered for traceability. A thermal shield cast iron block and a gunbarrel were analyzed. Their chemical compositions are given in Table 5. The chemical analysis focused on determining parent or target nuclei of long-lived radionuclides; therefore, the cobalt with its short (5-year) half-life was not included in the analysis.

TABLE 5

CHEMICAL COMPOSITION OF REACTOR PARTS

Chemical Element	Cast Iron from the Thermal Shield Block	Sample of Steel from the Gunbarrel Tip
Ti	0.012%	0.012%
V	0.004%	0.004%
Cr	0.156%	0.042%
Mn	0.248%	0.113%
Fe	98.8%	99.4%
Ni	140 ppm	130 ppm
Cu	0.288%	0.071%
Zn	0.009%	0.006%
Ga	0.002%	0.002%
Ge	0.004%	0.002%
Se	0.001%*	0.001%*
Sr	0.001%*	0.001%*
Y	0.001%*	0.001%*
Zr	0.004%	0.0
Nb	4.9 ppm	2.6 ppm
Mo	210 ppm	39 ppm

*Minimum detection limit.

Stainless steel (18-8), 3/4-in., Schedule 40 tubes were embedded front-to-rear in the side, top, and bottom thermal shields to provide cooling water for the shields. The front and rear shields were cooled by the process tubes. The induced activity of the thermal shield cooling tubes was calculated based on the fluence level and standard material specifications for 18-8 stainless steel. The calculated inventories for nickel-63, nickel-59, cobalt-60, molybdenum-93, and niobium-94 in the cooling tubes are included in the thermal shield inventories.

To account for the activity of the gunbarrels, the assumption was made that the inventory in the gunbarrels would be less than the thermal shield portion that they replace. This assumption is conservative for the radionuclides of interest as can be seen by inspection of the nickel, zirconium, niobium, and molybdenum composition values presented in Table 5.

The active portion of the thermal shields was considered to have the same length and height as the graphite stack. This assumption neglects the "corners" at each edge of the total shield; however, the neutron flux level in these corners is very low compared to the central portion of each shield face.

The inventory estimates for the thermal shields contain a conservatism similar to the graphite stack inventories. The concentrations, whether from measurements or calculated data, were applied to the entire active portion of the shield with no adjustment for lower neutron flux levels near the edges. The general method of determining the cobalt-60, nickel-63, and nickel-59 inventories was to consider the shields as eighteen slabs, each 1/4 in. thick. The measured values, as a function of distance into the shield from the DR Reactor data in Appendix A, were plotted on graph paper. From the graph a concentration for the desired radionuclide was determined for any specific 1/4-in. slab. By multiplying the concentration by the mass of the slab, the curies of a nuclide in each slab were determined. The sum of the curies over all the slabs gives the total shield inventory.

The estimated trace radionuclide inventories were calculated using a flux level of $10^{11}/cm^2$-s for the effective full-power days of operation and the impurity levels from the chemical analysis.

2.2.3 Process Tubes

The six older reactors used process tubes made of 1100 aluminum alloy. The chemical composition specification required a minimum of 99% aluminum. Other elements were allowed only to the maximum amounts listed in Table 6. These impurities do not generate significant quantities of radionuclides with long half-lives that are of concern. Gamma radiation spectrum measurement of aluminum samples that were present in the K Reactors for the entire operating life of the plant did not indicate measurable concentrations of aluminum-26.

TABLE 6

ALUMINUM ALLOY PROCESS TUBE IMPURITIES

Element	Maximum Amount
Cu	0.20%
Si + Fe	1.00%
Zn	0.10%
Mn	0.05%
Li	0.008%
Cd	0.003%
B + Co	0.001%
Others, each	0.05%

About 73% of the process tubes in the K Reactors are made of Zircaloy-2 and had been in use for 7 years prior to final shutdown. A chemical analysis of a Zircaloy-2 process tube was made to determine the possible elements that would produce long half-life nuclides. The analysis indicated 416 ppm nickel, 99.5% zirconium, and 14 ppm niobium. The only other element of concern is molybdenum, which is controlled by the purchase specification to be less than 50 ppm. Recent vendor reports for Zircaloy-2 purchased at Hanford indicate 25

ppm (minimum detectable) from Western Zirconium and 10 ppm (minimum detectable) from Wah Chang. To be conservative a value of 50 ppm was used for molybdenum.

The cladding used for the fuel elements contained 1% nickel, which combined with impurities in the cooling water to become part of the corrosion film inside the tube. A sample analysis was performed for both the film and the aluminum process tube material for several tubes at the DR Reactor (Appendix A). A comparable corrosion film analysis was not performed for the K Reactors because the activity of the K Reactor film is not expected to be significantly different since the same process water chemistry was used at all the reactors.

2.2.4 Reactor Control System

The reactor control system included the control rods, safety rods, and ball 3X system. The control rods, moved into and out of passages in the graphite core, controlled startup transients and power level during equilibrium operation. During later years, as the graphite stacks became distorted by growth and shrinkage, the control rod channels became distorted. Several "jointed" type rods were tested to allow the rod to follow the curved path of the channel. Also some testing was made of uncooled rods. All the original control rods were water cooled. No attempt was made to track the history of control rod replacement or short-time use of exotic designs.

The safety rods were located on top of the reactor. Electromagnets held the rods with just the rod tips in the top thermal shield. Only on reactor shutdown, automatic or intentional, were the safety rods dropped into the reactor proper. As the name implies, the safety rods were only a backup to the control rods. The tips of the safety rods became activated due to neutron streaming through the safety rod channels in the graphite stacks.

To back up the safety rods, a separate emergency shut-down system was provided. This was the ball 3X system, comprised of hoppers full of small

boron-steel balls that would automatically drain into the safety rod channels if the safety rods did not terminate the chain reaction after receiving a shut-down signal. Throughout the operating history of the Hanford reactors, the ball 3X systems were never required as the primary shut-down mechanism. However, the ball 3X systems were routinely tested manually to assure that all the balls in the hoppers would drain down into the channels in the graphite stack. During the later years of operation, as cracks and general shifting of the graphite stacks formed holes and ledges along the channels, balls would become trapped in the stack. These trapped balls caused a loss of reactivity which was overcome by adding more enriched fuel. The amount of enrichment needed to overcome the trapped balls was compared to the mass of balls in the reactor. For the DR Reactor, this equated to 72 pounds of balls. Measured data on the gamma radiation of a small number of irradiated balls were used to compute the amount of cobalt-60 that would be present in the trapped balls. Using this method, a cobalt-60 inventory of 110 curies was computed for the balls remaining in the graphite stack at the DR Reactor.

The inventory of the control rods and safety rod tips is assumed to be the same as the inventory of the thermal shield section (opening) that they replaced. Hence, the inventory of the rods is "included" in the thermal shield estimates. Prior to actual decommissioning a detailed characterization and radionuclide inventory of the control rods at each of the reactors should be made to determine the disposition of each rod on a case-by-case basis.

2.2.5 Biological Shield

The biological shield surrounds the thermal shield. The biological shields for the six older reactors were made of alternating laminated layers of steel and Masonite. As the single exception of the older reactors, the C Reactor has a 7-foot-thick, heavy-aggregate concrete top shield. Both K Reactors have heavy-aggregate concrete biological shields. While not especially intended as shields, all the reactors have massive concrete foundations.

Even though the biological shields are quite large in size and mass, they contain very little of the radionuclide inventory. The data presented in Appendix A indicate that less than 1% of total inventory of the thermal and biological shields combined is in the biological shield. Furthermore, of the inventory in the biological shields nearly all is in the first layer of steel. Considering the accuracy of the data in this document, the contribution of the biological shield cobalt-60, nickel-63, and nickel-59 may be treated as zero.

The amount of calcium-41 was calculated for the foundation and for the K Reactor shields for completeness. The calcium-41 was based on a calcium-40 level of 72,000 ppm for concrete and a flux level of $10^{10}/cm^2$-s.

2.3 ESTIMATES FOR REACTOR BLOCKS

The inventory for the reactor blocks is composed of estimates from the major components of the block: the graphite moderator stack, the thermal shield, the process tubes, and the control system, which includes the control rods, safety rods, and ball 3X systems. The inventory estimates will be tabulated at the end of the sections discussing the individual radionuclides.

2.3.1 Graphite Moderator Stack

Radionuclides in the graphite originate from the carbon, the cover gas, the cooling water (due to occasional tube leaks), and from the impurities present in the coke when it was processed into graphite blocks. The percentages of impurities present in the petroleum when it was baked into coke remained as the coke was further processed into graphite. The amount and configuration of impurities dictated the grade of graphite produced and its eventual suitability for reactor use. Initially the graphite was purified by heat only. Later, a chemical purification process was used to further reduce unwanted elements. The concentrations of radionuclides remaining in the graphite at each reactor reflect the source of the graphite, the grade of

graphite, and the history of operation. Tritium constitutes about half of the radioactivity present in the graphite. The other half is mainly carbon-14, nickel-63, cobalt-60, and chlorine-36.

2.3.1.1 Tritium

The amount of nitrogen-14 available as a target material in the graphite blocks was dictated by the concentrations in the original coke. The nitrogen-14 captures neutrons and becomes carbon-12 plus tritium. This is the dominant production reaction for tritium in the graphite stack (Appendix D). Tritium has a half-life of 12.3 years and emits low-energy beta particles.

Using the average value for tritium concentrations, shown in Table A-7 of Appendix A, yields a value of 6.8×10^6 pCi/g for the DR Reactor graphite for March 1977. Applying the central zone and reflector masses, scaling by the lower flux value assumed for the reflector, and, finally, decay-correcting to March 1985, an estimated inventory of 4900 curies of tritium is found for the DR Reactor stack. To determine the tritium inventories for the other old reactors, the ratio of the fluences is applied to the DR Reactor value of 6.8×10^6 pCi/g to obtain the other reactors' concentrations 12.2 years after shutdown. (The 12.2 years is the time between final shutdown of DR and the sample analysis time for data in Appendix A.) The value found for the tritium is then decay-corrected to March 1985 to be consistent with the inventory estimates presented in this document.

For the K Reactors a correction for the higher neutron flux level in the active zone, the greater mass of the active zone, and greater mass of the reflector is made in addition to the adjustments above for the older reactors.

No correction was made for the decay of tritium during the longer operating period of the reactors other than DR. This omission provides a conservatism in the estimates.

Table 7 shows the tritium inventory for the graphite stacks.

2.3.1.2 Carbon-14

Carbon-14 originates from both the nitrogen-14 (n, p) reaction and the (n, gamma) transmutation of carbon-13. The carbon-14 concentration in a graphite stack from the (n,p) reaction is predominantly a function of the available nitrogen in the graphite and cover gas. Samples of graphite from KW Reactor's tube-channel 1880 had a carbon-14 concentration similar to samples from DR Reactor (Appendix B). This similarity is probably due to differences in impurity levels, since a larger fraction of the graphite for K Reactors was chemically purified resulting in a lower impurity level. This lower impurity level could have offset the increased size and higher power levels of the K Reactors in determining carbon-14 inventories.

The average concentration of carbon-14 from the DR Reactor sampling (Appendix A) is 2.8×10^6 pCi/g for the active region of the reactor. Applying the adjustments discussed in Section 2.2.1, the inventory of carbon-14 for the DR Reactor is estimated to be 2990 for the active region and 176 for the reflector or 3200 curies total. The carbon-14 inventories for the other older reactors were estimated by using the ratios of the fluences compared to the DR Reactor.

The carbon-14 inventories for the K Reactors were based on the analysis data from the KW Reactor (Appendix B). The KE Reactor inventory was obtained by using the ratio of the fluences for KE and KW Reactors.

The carbon-14 inventories for the reactor graphite stacks are shown in Table 7.

2.3.1.3 Nickel

The original elements in the coke used to make the graphite influenced the amount of nickel available as a target material in the graphite. Nickel activation resulted in the generation of nickel-63 and nickel-59. Although a thermal purification process reduced the nickel in the original coke used to make the graphite in the older reactors, the nickel remaining in the graphite

appears to be proportional to the original concentration of the nickel impurity in the petroleum. For the K Reactor graphites, an additional chemical purification process significantly reduced the amount of nickel impurity present in the graphite. Nickel-62 constituted 3.6% of the total nickel present in the older reactor graphite. Upon capture of a neutron, nickel-62 becomes nickel-63 with a 100-year half-life.

Nickel-58 made up 68.3% of the nickel present in older reactor graphite. Neutron capture converts nickel-58 to nickel-59, with an 80,000-year half-life. It has been estimated that there are 0.008 curies of nickel-59 for each curie of nickel-63 (Appendix E). Other naturally occurring isotopes of nickel are of no interest since they do not produce radionuclides with long half-lives.

The average concentration of nickel-63 reported in Appendix A for the DR Reactor analysis is 8.4×10^4 pCi/g. Making the same corrections for active region and reflector as indicated for carbon-14 in Section 2.1.1.2, the total estimated inventory of nickel-63 in the DR Reactor graphite stack is 93 curies. Using the ratio of nickel-59 to nickel-63 derived in Appendix E, the estimated inventory of nickel-59 in the DR Reactor graphite stack is about one curie.

The nickel-63 and nickel-59 estimated inventories for the other reactors were determined by multiplying the ratio of the fluences and the average nickel content compared to the DR Reactor by the DR Reactor values above.

The K Reactors contained chemically purified graphite which resulted in a significantly lower nickel content. The nickel-63 and nickel-59 inventory estimates were made by adjusting the DR Reactor data for fluence and multiplying by the ratio of the average nickel content.

Table 7 shows the nickel inventory estimates for the reactor graphite stacks.

2.3.1.4 Chlorine-36

The amount of chlorine-35 available for transmutation to chlorine-36 depends upon the residual chlorine in the graphite after manufacture. At the time of construction of the early Hanford reactors (B, D, and F), the stacks were segregated by "purity" with respect to neutron absorbency. The higher grades of graphite (less neutron absorption) were placed in the center of the reactor and lower grades toward the outside. By the time the next generation of reactors (DR, C, and H) were constructed, an added process using carbon tetrachloride and freon was being used to purify the graphite during manufacture.

The amount of chlorine-36 is different for each reactor due to the varying amounts of each grade used during construction of the stack (Appendix C). The inventory for each reactor is shown in Table 7.

2.3.1.5 Cobalt-60

The amount of cobalt-60 present in the reactor graphite stacks is dependent upon the original impurity level of cobalt in the graphite. The purification process discussed earlier in Section 2.3.1 influenced the amount of cobalt-60 presently estimated for the graphite stack inventories.

Cobalt-60 has a half-life of 5.3 years, hence decays by several half-lives during the operation of the reactor. The estimated cobalt-60 inventory for the DR Reactor graphite was found by applying the volume adjustments for the active zone and the reflector and decay-correcting the average of the sample data in Appendix A to March 1985. The cobalt-60 inventories for the C and H Reactors were estimated by correcting the DR Reactor data for the difference in operating days and decay times since final shutdown. For the B, D, and F Reactors the same purification factor that was assumed for the nickel (Section 2.3.1.3) was applied, i.e., the B, D, F operation and decay time corrections and a factor of 2 to account for the higher impurity in the original graphite.

Similar corrections were made for the cobalt-60 in the KE and KW graphite stacks. In the case of the K Reactors the impurity correction factor is 0.04 rather than the 2 used for the B, D, and F graphites.

2.3.1.6 Calcium-41

The grade of graphite used to construct the reactor stacks influences the amount of calcium-41 that is produced during reactor operation. To determine the inventory of calcium-41 in each reactor required the same type of analyses performed for the chlorine-36 (Section 2.3.1.4). The computational method was to find the representative concentration of calcium for each zone of each reactor and then use the data set in Reference 1 to determine the calcium-41 produced.

2.3.1.7 Fission Products and Transuranics

For fission products to enter the graphite stack, both a fuel element failure and a simultaneous failure of the process tube are required, which would allow water and fission products to be carried into the graphite. The amount of fission products remaining in the graphite after nine years of decay would add a few curies to the total inventory of the reactor. As indicated in Table 7, the inventory from mixed fission products remaining in the graphite stacks is made up of plutonium-239, strontium-90, and cesium-137. The estimates were calculated from the DR Reactor data (Appendix A) and applied to the other reactors without correction or adjustment.

Table 7 shows the present estimated radionuclide inventory in the reactor graphite stacks.

TABLE 7

ESTIMATED RADIONUCLIDE INVENTORY
IN THE REACTOR GRAPHITE
AS OF MARCH 1, 1985

Radio-nuclide	Reactor Graphite (curies)							
	B	C	D	DR	F	H	KE	KW
^{3}H	8300	8900	7700	4900	5800	5500	30000	27000
^{14}C	4500	4500	4300	3200	3700	3500	7000	6700
^{60}Co	100	60	90	30	70	40	5	5
^{63}Ni	180	28	280	95	190	120	11	15
^{59}Ni	1	1	2	1	2	1	-	-
^{36}Cl	42	12	34	26	33	17	54	52
^{239}Pu	1	1	1	1	1	1	1	1
^{241}Am	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
^{90}Sr	10	10	10	10	10	10	10	10
^{137}Cs	30	30	30	30	30	30	30	30
^{133}Ba	32	1	34	10	26	11	1	1
^{152}Eu	40	40	40	40	40	40	40	40
^{154}Eu	20	20	20	20	20	20	20	20
^{41}Ca	190	14	150	90	140	54	1	5

2.3.2 Thermal Shield

Table 8 gives the DR Reactor distribution of cobalt-60 and nickel-63 in the right side thermal shield as a function of distance from the inner face of the shield. This distribution is representative of all the thermal shield faces. The curies were obtained by multiplying each activity by the mass of the slab, 4.46×10^6 grams.

Total curies of cobalt-60, nickel-63 and nickel-59 for the DR Reactor thermal shield sections are shown in Table 9. The table reflects the different masses of the front and rear shields.

TABLE 8

DISTRIBUTION OF COBALT-60 AND NICKEL-63 IN THE DR REACTOR
RIGHT SIDE THERMAL SHIELD
AS OF MARCH 1, 1985

Distance from Inner Face (in.)	Cobalt-60 (pCi/g)	Nickel-63 (pCi/g)	Cobalt-60 (Ci)	Nickel-63 (Ci/cm)
0.25	3.4×10^7	2.4×10^6	153	10.5
0.50	2.5×10^7	1.7×10^6	114	7.7
0.75	1.9×10^7	1.3×10^6	85	5.7
1.0	1.4×10^7	9.4×10^5	63	4.2
1.25	1.1×10^7	7.0×10^5	47	3.1
1.5	7.8×10^6	5.1×10^5	35	2.3
1.75	5.8×10^6	3.8×10^5	26	1.7
2.0	4.2×10^6	2.8×10^5	19	1.2
2.25	3.2×10^6	2.0×10^5	15	0.9
2.50	2.4×10^6	1.5×10^5	11	0.7
2.75	1.8×10^6	1.1×10^5	8	0.5
3 thru 8	---	---	24	1.4
		Total curies	600	39.9

TABLE 9

COBALT-60, NICKEL-63, AND NICKEL-59 IN THE DR THERMAL SHIELD
MARCH 1, 1985

Shield Section	Cobalt-60 Curies	Nickel-63 Curies	Nickel-59 Curies
Right	600	40	0.32
Left	600	40	0.32
Top	600	40	0.32
Bottom	600	40	0.32
Front*	780	52	0.42
Rear*	780	52	0.42
TOTAL	3960	264	2.1

*Corrected for volume and mass.

The cobalt-60 and nickel-63 inventories for the reactor thermal shields were estimated by comparing the DR Reactor sample data (Appendix A) to the other reactors. Where sample data were not available, e.g., nickel-59, the inventories were calculated based on the ratio of nickel-59 to nickel-63 described in Appendix E. To determine the DR Reactor shield inventory, the sample data were plotted on graph paper to find the specific activity as a function of distance from the inner face of the shield. By considering the shield to be made up of a series of 1/4-in. thick slabs with the same height and length as the adjacent graphite stack, the total curie inventory for each slab could be estimated.

The cobalt-60 inventory estimate for the DR Reactor was adjusted for the different operating times and decay times for the other older reactors. The K Reactors were additionally adjusted for the greater mass of the shields. Nickel-63 and nickel-59 inventories were adjusted only for the different operating times and masses since the difference due to varying decay times is negligible.

The thermal shield cooling tubes contribute significantly to the nickel-63 and nickel-59 inventories since the stainless steel used for the tubes was 8 % nickel. The inventory estimates for the radionuclides in the cooling tubes were calculated using a flux level of 10^{11} neutrons/cm^2-s for the total equivalent operating days of each reactor. The K Reactors required a further adjustment to account for the increased number and length of the tubes.

Table 9 shows the estimated inventories for the reactor thermal shields and cooling tubes.

TABLE 10

RADIONUCLIDE INVENTORIES FOR THE REACTOR THERMAL SHIELDS
AS OF MARCH 1, 1985

Reactor	Cobalt-60 (Curies)	Nickel-63 (Curies)	Nickel-59 (Curies)
B shield	8600	380	3
cooling tubes	90	460	4
C shield	9800	380	3
cooling tubes	90	460	4
D shield	7300	370	3
cooling tubes	80	440	4
DR shield	4000	260	2
cooling tubes	60	320	3
F shield	4800	310	2
cooling tubes	70	370	4
H shield	4200	300	2
cooling tubes	70	350	3
KE shield	17200	550	4
cooling tubes	260	650	5
KW shield	14300	510	4
cooling tubes	220	620	5

2.3.3 Process Tubes

Radionuclides associated with the process tubes are from two sources: activation of the tube materials and film deposits on the inner tube wall from corrosion products. Samples of three process tubes were taken from the DR Reactor for analysis (Appendix A). The analyses were performed on the process tube material and on the film layer. In order to apply the DR Reactor sample data to the entire reactor or to the other Hanford reactors, some clarification is necessary.

The induced activity of the process tube material varies from front to rear of the reactor according to the neutron flux distribution. Ignoring local effects of control rods, the neutron flux distribution may be approximated by a cosine over the length of interest. That is, the flux level is a maximum at the center of the reactor and drops by several orders of magnitude several feet past the end of the fuel charge.

The activity of the film layer inside the process tubes starts at a low value near the front face end of the fuel charge (upstream end) and reaches a maximum value past the rear face end of the fuel charge (downstream end). Since the reactors in this document used "single pass" cooling in which cooling water was not recirculated, the activity along the process tube film maintained the "S" shape distribution throughout the life of the tube.

The DR Reactor process tube samples were obtained about one-fourth of the fuel charge length from the downstream end. At this position the neutron-induced activity in the process tube was lower than at the center of the tube but overall was nearly average for the entire length of tube. The activity of the film was less than the maximum concentration, but provides a conservative estimate when applied to the entire tube length, since film upstream of the sample point would be of lower activity.

The DR Reactor process tube film samples were apparently not analyzed for the nickel isotopes in this document. The fuel element cladding contained about 1% nickel. Some of the cladding material combined with the impurities in the process water and became part of the film on the process tubes. For the older reactors the nickel-63 inventory of the process tubes was estimated to be about 10 curies. If the nickel-63 is 10 curies, then the nickel-59 inventory is about 0.1 curies. Since a film analysis was not performed for the K Reactors, a factor of 1.5 was applied to the older reactor values to account for the greater number of process tubes and the greater length of the tubes.

A chemical analysis was made of a piece of unirradiated Zircaloy-2 process tube to identify elements that would produce long half-life nuclides by neutron activation. The elements of interest are nickel, zirconium, niobium, and molybdenum. The analysis results were stated earlier in Section 2.2.3. Calculations were made for the estimated inventories of zirconium-93, nickel-63, and nickel-59 in the Zircaloy-2 process tubes for the K Reactors. The estimated inventories for molybdenum-93 and niobium-94 were calculated from the data set given in Reference 1 with adjustment for the actual exposure time of the Zircaloy-2 process tubes in the K Reactors.

A summary of the estimated radionuclide inventory for the reactor process tubes is given in Table 11.

TABLE 11

ESTIMATED RADIONUCLIDE INVENTORY IN THE
REACTOR PROCESS TUBES

Radio-Nuclide	Reactor (Curies)							
	B	C	D	DR	F	H	KE	KW
^{60}Co	300	350	270	200	210	200	190	170
^{59}Ni	0.1	0.1	0.1	0.1	0.1	0.1	13	11
^{63}Ni	10	10	10	10	10	10	1700 17	1500 15
^{90}Sr	0.2	0.2	0.2	0.2	02	0.2	0.3	0.3
^{93}Mo	--	--	--	--	--	--	0.2	0.2
^{93}Zr	--	--	--	--	--	--	11	10
^{152}Eu	1.6	1.7	1.5	1.3	1.4	1.3	2	2
^{154}Eu	1.2	1.3	1.1	0.9	1.0	1.0	1.6	1.6

2.3.4 Reactor Control System

The vertical safety rods, horizontal controls rods, and ball 3X safety system make up the reactor control system. Only a small segment of each control rod was exposed to the reactor's neutron fluence. Additionally, only the reactor entry ports of the ball system were exposed. The degree to which these exposed portions became radioactive depended on the component's chemical composition. The radionuclide inventories of the vertical and horizontal control rods are assumed to be equal to the amounts of thermal shield removed by the penetrations. See Section 2.2.4.

When the ball 3X system balls were dropped into the reactor, a small number of balls became trapped in the openings of the graphite and could not be removed. These trapped balls caused a reactivity loss which was overcome by adding more enriched fuel. The amount of enrichment needed was equated to the mass of the balls in the reactor. For DR Reactor, this equated to 72 pounds of balls. Measured data on the gamma emission of a small number of irradiated balls were used to compute the amount of cobalt-60 that would be present in the trapped balls. Using this method, a cobalt-60 inventory of 110 curies was computed for the balls remaining in the DR Reactor core. The DR Reactor had the greatest amount of enriched fuel added to compensate for the balls in the stack. As a conservative measure 110 curies of cobalt-60 were added to each of the other seven reactor inventories.

2.3.5 Trace Radionuclides

Impurities in the materials exposed to the neutron flux became activated during reactor operation. While the contribution of these activated impurities is small compared to the major constituents considered above, calculations were performed to indicate the amounts of trace radionuclides that may be present in various reactor components. The calculations in most cases were based on the data set provided in Reference 1 with neutron flux levels of 10^{14}, 10^{11}, and 10^{10} assumed for the graphite stacks, the

thermal shields, and the biological shields respectively. In cases where an impurity level had not been chemically analyzed, the suggested value given in Reference 1 was used for the calculations. In the case of technicium-99 production from neutron activation of molybdenum-98, a manual calculation was made using basic activation formula. No corrections or adjustments were made to determine the trace radionuclides for each reactor. Instead, the inventories were grouped by the older reactors and the K reactors.

Tables 12 and 13 show the impurity level of the parent isotope, the radionuclide produced, and the calculated curie content of the reactor rcomponent.

TABLE 12

TRACE RADIONUCLIDE INVENTORIES FOR
B, C, D, DR, F AND H REACTOR COMPONENTS

Component	Parent Nuclide and Concentration (ppm)		Radionuclide Formed	Curies Per Component
Graphite stack	Niobium	0.15	^{94}Nb	0.3
Thermal shield	Silver	0.35	^{108}Ag	0.03
	Niobium	4.9	^{94}Nb	0.02
	Molybdenum	210	^{93}Mo	0.04
			^{99}Tc	0.002
Concrete Base	Calcium	72000	^{41}Ca	2
Top biolog- ical shield (C Reactor only)	Calcium	72000	^{41}Ca	2

TABLE 13

TRACE RADIONUCLIDE INVENTORIES FOR

KE AND KW REACTOR. COMPONENTS

Component	Parent Nuclide and Concentration (ppm)		Radionuclide Formed	Curies Per Component
Graphite stack	Niobium	0.15	^{94}Nb	1.1
Thermal shield	Silver	0.35	^{108}Ag	0.04
	Niobium	4.9	^{94}Nb	0.03
	Molybdenum	210	^{93}Mo	0.06
			^{99}Tc	0.003
Zircaloy-2 Process Tubes	Niobium	14	^{94}Nb	0.6
	Molybdenum	50	^{93}Mo	0.2
			^{99}Tc	0.03
Concrete Base and Biological Shield	Calcium	72000	^{41}Ca	15

3.0 FUEL STORAGE BASINS

Each reactor building contains an irradiated fuel storage basin. The basin
served as a collection, storage, and transfer facility for the fuel elements
discharged from the reactor. A typical reactor fuel storage basin consists of
the fuel element pickup chute area, the storage area, and the transfer area.
Irradiated fuel elements were sorted in the pickup chute area and hand-tonged
into storage buckets, and then transferred by an overhead monorail system to
the storage area where they were held for the decay of short-lived
radionuclides. Following the storage period, the buckets of fuel elements
were moved by the overhead monorail system to the transfer area, where they
were placed in railroad cask cars for transport to the chemical reprocessing
facilities in the Hanford 200 Area.

During the past two years work has been initiated to clean up and stabilize
the basins prior to decommissioning. The D and DR basins were washed down,
the sludge was removed and taken to the 200 Area waste disposal site, and an
asphalt emulsion (ATCO 1840) was applied to the floor and walls to fix any
remaining contamination. At the B and C Reactor basins, the sludge was moved
to the lower part of the fuel transfer pits and shielded with a cap, and the
walls above the sludge in the pits, and the walls and floors of the basins
were coated with the asphalt emulsion. The K Reactor storage basins are still
in operation, storing irradiated fuel from the N Reactor. The F and H Reactor
fuel storage basins were backfilled with soil in 1970. The soil covers the
sludge and miscellaneous equipment that were in the basins at the time.

3.1 METHODS AND SOURCES FOR FUEL STORAGE BASINS

The radionuclides in the fuel storage basins originated from process tube
scale and from failed fuel elements that were discharged into the basins.
Over the years sludge accumulated on the floors of the basins. The sludge is
the primary source for the basin inventory.

Sample data were obtained from the sludges prior to removal and estimates of residual inventories were made from survey data taken after cleanout. As the B Reactor basin had the highest overall inventory prior to cleanout, that inventory was used as an estimate for the F and H Reactor basins. The K Reactor basins will be cleaned prior to decommissioning, hence, the inventory of the DR Reactor basin is applied to the K Reactor basins.

The water was removed from the B and C fuel storage basins. The contaminated sludge was pumped into the transfer area pits. The basin walls were washed down using high pressure water and then coated with an asphalt-based emulsion to fix the residual contamination to the surface. Additional characterization of the sludge in the transfer area pits may be required prior to final disposition.

The status of the D and DR basins is the same as the B and C basins, except that the sludge was removed from the basins and buried in the 200 Area low-level waste burial ground.

KE and KW fuel storage basins are still in use to store N Reactor fuel. It is assumed that these basins will be drained and the sludge will be removed, packaged, and shipped for disposal. The basins' floors and walls will be cleaned and the contamination on the surfaces fixed.

After the shutdown of F and H Reactors, the irradiated fuel was shipped to the 200 Area, the storage basins' water was pumped down to within a few feet above the basin floor and a detailed radiological and visual survey was conducted of the basins. After the completion of the survey and removal of identified high-dose-rate materials, the two basins were backfilled with soil in 1970. Irradiated spacers, process tubing, buckets, and miscellaneous reactor hardware were not removed prior to backfilling. Additionally, there is a possibility that one or more low-dose-rate irradiated fuel elements

were overlooked and buried under the 20 feet of earth backfill. If the soil fill is removed from the F and H basins, the fuel elements, if any, will be shipped to the 200 Areas for final disposition and the sludge mixed with the soil will be packaged and shipped to the 200 Area low-level waste burial site. If the decision is made not to remove the soil from the basins, the sludge inventory from the B Reactor basin (highest inventory of all the basins) and a conservative inventory of possible fuel elements should be added. The calculated inventory of five fuel elements is shown in Table 14.

TABLE 14

ESTIMATED INVENTORY OF FIVE SPENT FUEL
ELEMENTS (Assumes 15-year-old Fuel)

Radionuclide	Half-life (years)	Curies
^{90}Sr	29	40
^{137}Cs	30.2	40
^{152}Eu	13.4	0.0003
^{238}Pu	87.7	0.02
^{239}Pu	24110	1.2
^{240}Pu	6537	0.3
^{241}Pu	14.7	10
^{85}Kr	10.7	2
^{151}Sm	90	0.4
^{113}Cd	14.6	0.002
^{94}Nb	20000	0.0008
^{79}Se	65000	0.0002
^{107}Pd	6500000	0.00002
^{241}Am	432	0.4
^{99}Tc	213000	1
^{93}Zr	1500000	0.002
U_{238}	4500000000	0.006

3.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR FUEL STORAGE BASINS

It is estimated that each of the basins at B, C, F, and H Reactors contains 50,000 kg of sludge. The radionuclide inventories for the fuel storage basins that contain sludge were calculated by multiplying the 50,000 kg by measured nuclide concentrations. The inventories for D and DR Reactor basins were determined from surveys taken after cleanout of the basins. The KE and KW Reactor basins are still in use to store irradiated fuel from N Reactor. It is assumed that the K Reactor basins will be cleaned at least to the present levels at the D and DR basins prior to decommissioning.

During the cleanout of the D and DR Reactor basins five irradiated fuel elements (two at D and three at DR) were found in the sludge on the basin floors. The layaway procedures used at the D and DR Reactors were much the same as used at the F and H Reactors. Since fuel elements were found at D and DR, it must be assumed that elements could also be present in the F and H Reactor basins. To provide a basis for the EIS evaluation, the radionuclide inventory for five fuel elements are listed.

Should the decision be made not to remove the earthen fill from the basins to search for spent fuel, the estimated inventories for F and H should be adjusted to add the inventory of the radionuclides identified as part of the fuel elements and estimated sludge.

3.3 ESTIMATES FOR FUEL STORAGE BASINS

Based on the information stated, the fuel storage basin estimated inventories are listed in Table 15.

TABLE 15

FUEL STORAGE BASIN INVENTORY
CURIES AS OF MARCH 1, 1985

Basin	^{59}Ni	^{60}Co	^{63}Ni	^{90}Sr	^{137}Cs	^{152}Eu	^{154}Eu	^{238}U	^{239}Pu	^{241}Am	^{238}Pu
B	0.5	11	60	14	16	1.4	4.2	0.009	1.6	0.5	0.075
C	0.16	16	16	7	6	4	7	0.004	1.5	0.5	0.075
D	0.002	0.05	0.27	0.06	0.12	0.02	0.007	--	0.024	0.008	--
DR	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
F*	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
H*	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
KE	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
KW	0.01	0.23	1.25	0.29	0.81	0.23	0.05	--	0.024	0.008	--
TOTAL											

*Assumes that F and H basins are cleaned to same level as the DR basin. If
soil is left in the basins, assume B basin sludge inventory and add fuel
elements (see text).

4.0 CONTAMINATED AND ACTIVATED PORTIONS OUTSIDE THE REACTOR BLOCK AND FUEL STORAGE BASIN

There are reactor components and numerous facilities that are radioactively contaminated but not included in the previously discussed inventories. These components or locations are estimated to contain less than 5% of the total reactor facility inventory; therefore, exact inventories were not estimated. As examples, the soil columns under a fuel storage basin and reactor may be contaminated. These areas have not been characterized, but the inventory is estimated to be low when compared to the total inventory for the reactor facility.

While not within the stated scope of this document, several ground disposal facilities in the near vicinity of the reactor buildings were examined. If the in situ option for decommissioning the reactor were chosen, nine ground disposal facilities would be included in the 210-ft radius of the earthen mound. The affected facilities are the 116-B-3 Pluto Crib, the 116-B-4 Dummy Decontamination Drain, the 116-D-6 105-D Cushion Corridor French Drain, the 116-F-11 105-F Cushion Corridor French Drain, the 116-KE-1 115 Crib, the 116-KE-3 105-KE Storage Basin Drain, the 116-KW-1 115 Crib, the 116-KW-2 105-KW Storage Basin Drain, and the 105-C Ball 3X Silos. The calculated inventories for these facilities are included in Appendix F for information.

4.1 METHOD AND SOURCES FOR PORTIONS OUTSIDE THE REACTOR BLOCK AND BASINS

Radiation surveys of reactor building areas other than the reactor blocks and fuel storage basins indicated general radiation levels of less than 1 mrem/hr to 5 mrem/hr with isolated areas reading up to 10 mrem/hr. The areas of the survey included the front and rear work areas of the reactor, the pipe tunnels, the outer rod room, and other miscellaneous access areas within the reactor buildings.

4.2 CALCULATION ADJUSTMENTS AND ASSUMPTIONS FOR PORTIONS OUTSIDE THE REACTOR BLOCK AND BASINS

There were no inventory estimates made of the radionuclides that may be present in the form of contamination in reactor building spaces outside the reactor blocks and fuel storage basins. As noted in Section 4.1, the dose rates measured during a radiological survey were quite small.

5.0 SUMMARY OF RADIONUCLIDE INVENTORIES

The inventory estimates presented in this document are summarized in Tables 16-23 for each of the surplus reactor facilities. The total radionuclide inventory for a reactor facility ranges from 13,000 curies to just over 58,000 curies for the two reactor types, as shown by the tables.

The quantities stated in the inventories are conservative, as the methodology was designed to overestimate the radionuclides in the facilities. As examples of the conservatism, sample points in the thermal shield and the process tubes were selected for higher than average concentrations for a component.

The thermal shield calculations are based on the samples measured toward the center of the right side shield. Even though the samples would represent a higher fluence area of the shield, therefore a higher concentration of radionuclides, the same concentration of radionuclides was assumed for the entire shield.

The same type of conservatism was included in the process tube estimates. Samples were taken in the central zone of the reactor and the measured radionuclide inventory was applied to the entire reactor without accounting for the much lower fluence in the tube ends and in the fringe regions of the reactor.

To be certain that some large source had not been overlooked and that the correct areas of the facilities were included, an independent technical review of the inventories was performed by persons knowledgeable in the field but not directly involved in the estimates. The suggestions and comments from the technical review were incorporated as appropriate into this final document.

TABLE 16

RADIONUCLIDE INVENTORY ESTIMATE
B REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	8300	--	--	--	--	--
^{14}C	4500	--	--	--	--	--
^{41}Ca	190	--	--	--	2	--
^{60}Co	100	8690	300	110	--	11
^{59}Ni	1	7	0.1	--	--	0.5
^{63}Ni	180	840	10	--	--	60
^{36}Cl	42	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	14
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	16
^{152}Eu	40	--	1.6	--	--	1.4
^{154}Eu	20	--	1.2	--	--	4.2
^{238}U	--	--	--	--	--	0.009
^{238}Pu	--	--	--	--	--	0.075
^{239}Pu	1	--	--	--	--	1.6
^{241}Am	0.3	--	--	--	--	0.5

TABLE 17

RADIONUCLIDE INVENTORY ESTIMATE
C REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	8900	--	--	--	--	--
^{14}C	4500	--	--	--	--	--
^{41}Ca	14	--	--	--	4	--
^{60}Co	60	9890	350	110	--	16
^{59}Ni	--	7	0.1	--	--	0.16
^{63}Ni	28	840	10	--	--	16
^{36}Cl	12	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	7
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	6
^{152}Eu	40	--	1.7	--	--	4
^{154}Eu	20	--	1.3	--	--	7
^{238}U	--	--	--	--	--	0.004
^{238}Pu	--	--	--	--	--	0.075
^{239}Pu	1	--	--	--	--	1.5
^{241}Am	0.3	--	--	--	--	0.5

TABLE 18

RADIONUCLIDE INVENTORY ESTIMATE
D REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	7700	--	--	--	--	--
^{14}C	4300	--	--	--	--	--
^{41}Ca	150	--	--	--	2	--
^{60}Co	90	7380	270	110	--	0.05
^{59}Ni	2	7	0.1	--	--	0.002
^{63}Ni	280	810	10	--	--	0.27
^{36}Cl	34	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	0.06
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	0.12
^{152}Eu	40	--	1.7	--	--	2
^{154}Eu	20	--	1.2	--	--	0.007
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

TABLE 19

RADIONUCLIDE INVENTORY ESTIMATE
DR REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	4900	--	--	--	--	--
^{14}C	3200	--	--	--	--	--
^{41}Ca	90	--	--	--	2	--
^{60}Co	30	4060	200	110	--	0.23
^{59}Ni	1	5	0.1	--	--	0.01
^{63}Ni	95	580	10	--	--	1.25
^{36}Cl	26	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	0.29
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	0.81
^{152}Eu	40	--	1.3	--	--	0.23
^{154}Eu	20	--	0.9	--	--	0.05
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

TABLE 20

RADIONUCLIDE INVENTORY ESTIMATE
F REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	5800	--	--	--	--	--
^{14}C	3700	--	--	--	--	--
^{41}Ca	140	--	--	--	2	--
^{60}Co	70	4870	210	110	--	0.23
^{59}Ni	2	6	0.1	--	--	0.01
^{63}Ni	190	680	10	--	--	1.25
^{36}Cl	33	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	0.29
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	0.81
^{152}Eu	40	--	1.4	--	--	0.23
^{154}Eu	20	--	1.0	--	--	0.05
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

TABLE 21

RADIONUCLIDE INVENTORY ESTIMATE
H REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	5500	--	--	--	--	--
^{14}C	3500	--	--	--	--	--
^{41}Ca	54	--	--	--	2	--
^{60}Co	40	4270	200	110	--	0.23
^{59}Ni	1	5	0.1	--	--	0.01
^{63}Ni	120	650	10	--	--	1.25
^{36}Cl	17	--	--	--	--	--
^{90}Sr	10	--	0.2	--	--	0.29
^{93}Zr	--	--	--	--	--	--
^{93}Mo	--	0.04	--	--	--	--
^{94}Nb	0.3	0.02	--	--	--	--
^{99}Tc	--	0.002	--	--	--	--
^{108}Ag	--	0.03	--	--	--	--
^{137}Cs	30	--	--	--	--	0.81
^{152}Eu	40	--	1.3	--	--	0.23
^{154}Eu	20	--	1.0	--	--	0.05
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

TABLE 22

RADIONUCLIDE INVENTORY ESTIMATE
KE REACTOR SUMMARY
AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio-Shield	Storage Basin
^{3}H	30000	--	--	--	--	--
^{14}C	7000	--	--	--	--	--
^{41}Ca	1	--	--	--	15	--
^{60}Co	5	17500	190	110	--	0.23
^{59}Ni	--	9	13	--	--	0.01
^{63}Ni	11	1200	1700	--	--	1.25
^{36}Cl	54	--	--	--	--	--
^{90}Sr	10	--	0.3	--	--	0.29
^{93}Zr	--	--	11	--	--	--
^{93}Mo	--	0.06	0.2	--	--	--
^{94}Nb	1.1	0.03	0.6	--	--	--
^{99}Tc	--	0.003	0.03	--	--	--
^{108}Ag	--	0.04	--	--	--	--
^{137}Cs	30	--	--	--	--	0.81
^{152}Eu	40	--	2	--	--	0.23
^{154}Eu	20	--	1.6	--	--	0.05
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

TABLE 23

RADIONUCLIDE INVENTORY ESTIMATE

KW REACTOR SUMMARY

AS OF MARCH 1, 1985

Radio nuclide	Component (Curies)					
	Graphite Stack	Thermal Shield	Process Tubes	Control System	Bio- Shield	Storage Basin
^{3}H	27000	--	--	--	--	--
^{14}C	6700	--	--	--	--	--
^{41}Ca	5	--	--	--	15	--
^{60}Co	5	14500	170	110	--	0.23
^{59}Ni	--	9	11	--	--	0.01
^{63}Ni	15	1100	1500	--	--	1.25
^{36}Cl	52	--	--	--	--	--
^{90}Sr	10	--	0.3	--	--	0.29
^{93}Zr	--	--	10	--	--	--
^{93}Mo	--	0.06	0.2	--	--	--
^{94}Nb	1.1	0.03	0.6	--	--	--
^{99}Tc	--	0.003	0.03	--	--	--
^{108}Ag	--	0.04	--	--	--	--
^{137}Cs	30	--	--	--	--	0.81
^{152}Eu	40	--	2	--	--	0.23
^{154}Eu	20	--	1.6	--	--	0.05
^{238}U	--	--	--	--	--	--
^{238}Pu	--	--	--	--	--	--
^{239}Pu	1	--	--	--	--	0.024
^{241}Am	0.3	--	--	--	--	0.008

6.0 HAZARDOUS MATERIALS

Several materials considered to be hazardous were identified and inventoried on a recent inspection of the reactor facilities (Appendix G). Materials identified were lead, mercury, friable asbestos, and cadmium. The mercury and some of the asbestos have already been removed. Except for some of the lead, the other materials will be removed as necessary for disposal elsewhere.

The amounts of lead in the reactor facilities are divided into removable and nonremovable estimates. The removable lead actually fits two categories:
- Removal requires the assistance of some standard mechanical lifting device such as a fork lift; and
- Removal requires special dismantlement of the leaded assembly and building structure.

The lead considered nonremovable is "locked up" in the reactor block and would require actual dismantling of the blocks to gain access for its removal. The lead was exposed to a neutron flux and is, therefore, considered to be irradiated. The long-lived radionuclides, silver-108m, manganese-54, and bismuth-210m, have an estimated inventory of less than 1 curie for all eight reactors.

The lead external to the eight reactor blocks has low levels of surface contamination with an estimated inventory of less than 1 curie of mixed radionuclides.

The disposition of hazardous materials will be accomplished in accordance with applicable regulations at the time of decommissioning and will be addressed in a safety hazards assessment prior to initiating actual decommissioning work. In addition, definitive engineering and decommissioning and radiological work procedures will provide explicit instructions for control and disposal of all materials determined to be hazardous during the actual decommissioning effort.

7.0 REFERENCE

1. L. D. Felstead and P. B. Woollam, <u>An Assessment of all Known Isotopes to Determine Which Might be Important in the Decommissioning of Thermal Nuclear Reactors</u>, TPRD/B/0386/N84, January 1984.

APPENDIX A

REACTOR CHARACTERIZATION

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APPENDIX A

CHARACTERIZATION OF THE RADIOACTIVITY REMAINING IN THE
SURPLUS PRODUCTION REACTORS AT HANFORD

A.1 REACTOR SAMPLING

During 1976-1977, four sample holes were drilled into the DR Reactor block.
(See Figure A-1.) The sample holes were drilled through the biological and
thermal shielding and into the graphite stack. Sampling of the DR Reactor was
intended to establish the following:

o Radionuclide concentrations within the DR Reactor biological and thermal
  shields, and the graphite stack.

o In-place reactor dose rates at selected locations.

o Distribution of radionuclide concentrations in the thermal and biological
  shields with respect to depth of penetration.

In addition to the sampling and radiological monitoring performed at the DR
Reactor, in-place dose rate measurements were taken of selected process tubes
in the F and H Reactors.

Radioanalytical results of process tube sampling performed in 1967 at the
DR Reactor are also reported along with decay corrections. In addition,
samples had been taken of several gunbarrels from the C Reactor in 1967. The
results of radionuclide analyses of these samples are also reported.

A.2 SUMMARY OF SAMPLING RESULTS

Reactor sampling at the experimental facility locations (X-levels) of the
DR Reactor was initiated on December 28, 1976. Sampling of the first sample
hole, A, was completed on January 28, 1977. Drill shaving samples were taken
of the biological shield. A continuous core sample of the thermal shield was
taken and segmented for radionuclide analyses. Two graphite samples were
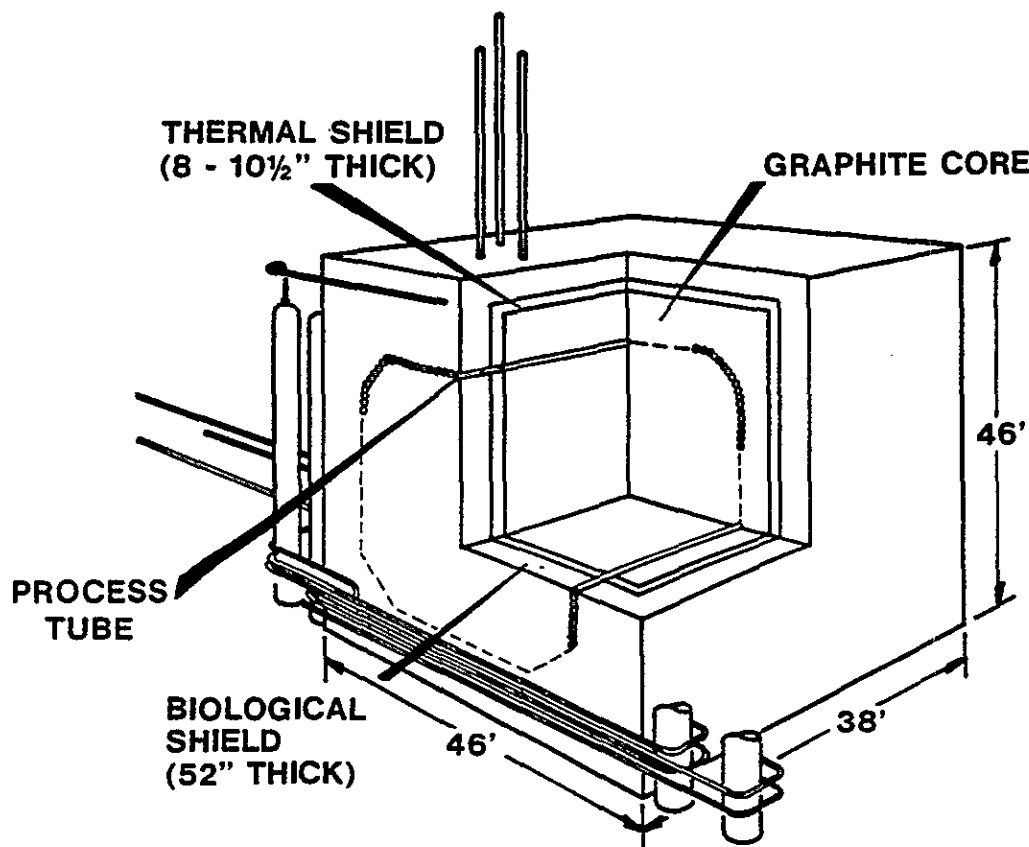
Figure A-1. Reactor Block Construction.
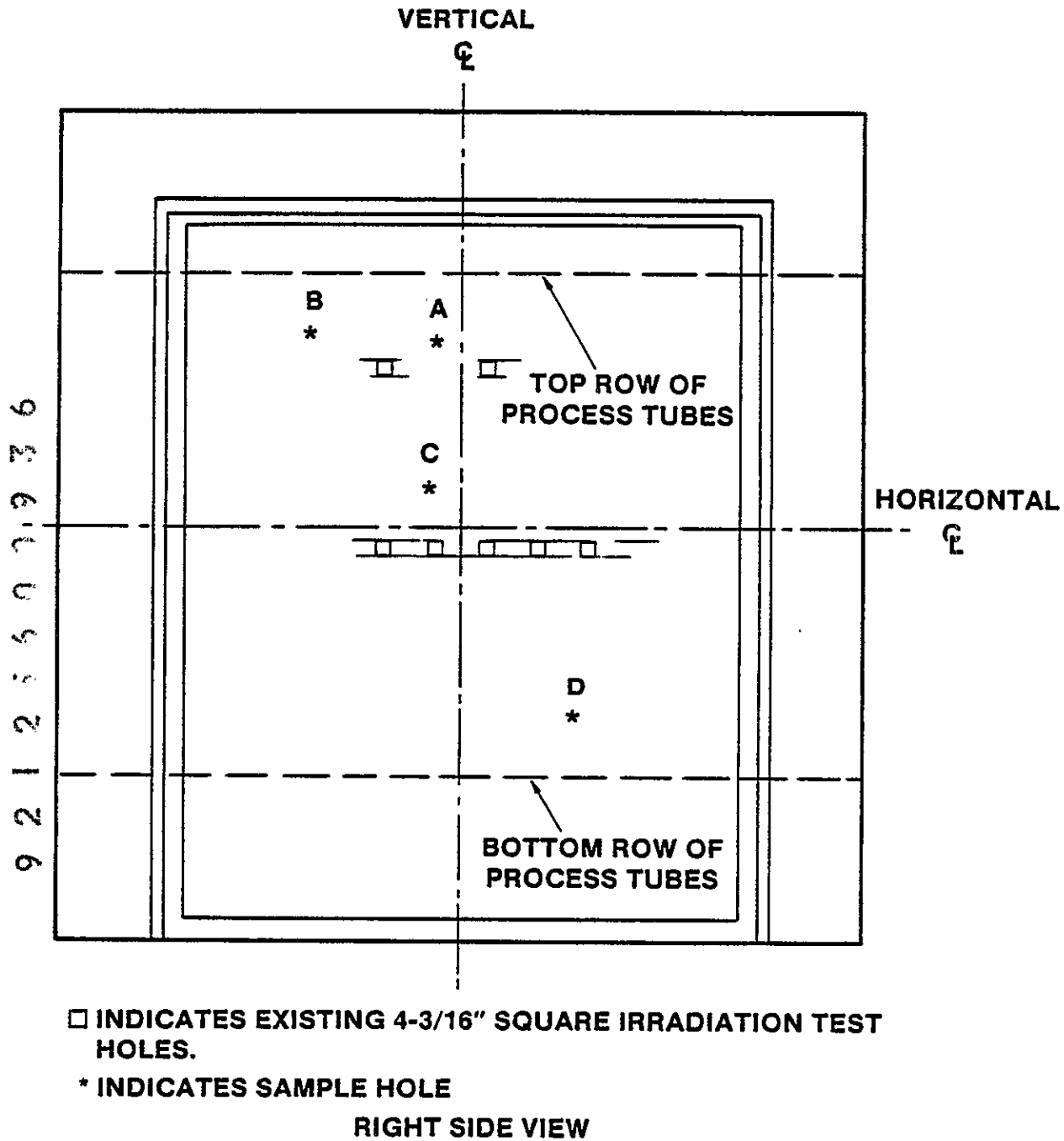
Figure A-2. Approximate Sample Hole Locations at the DR Reactor

taken in the reflector area and five graphite samples were taken in the active portion of the reactor to a total distance of 33 feet, which is 6 feet from the reflector area at the opposite side of the reactor. The indicated dose rate on the 33-foot sample was 20 mR/hr. All other graphite samples were less than 2 mR/hr.

By March 14, 1977, a second sample hole, B, had been completed, and the biological and thermal shield samples of sample hole C had been taken. At this point, the sampling was temporarily terminated and the drilling equipment placed on standby until additional funding could be obtained to complete the work. Additional funding became available in late August 1977. The sampling work was restarted on August 29 and completed on September 22, 1977. Selected samples were submitted for radioisotopic analyses.

Some general conclusions with respect to the radioactivity in the DR Reactor are as follows:

o In-place dose rates of selected process tubes average 50 R/hr in the vicinity of the gunbarrel tips and drop down to 4 R/hr 15 feet into the process tube from the nozzle.

o In-place dose rates at the outer surface of the thermal shield were approximately 2 R/hr at the sample hole locations.

o The maximum in-place dose rate measured was approximately 100 R/hr.

o Approximately 90 percent of the biological shield radioactivity is contained in the inner 1-3/4-inch layer of steel closest to the thermal shield.

o Approximately 90 percent of the total biological and thermal shield radioactivity is in the inner 2-inch portion of the thermal shield; 99 percent of the activity is in the inner 4-inch portion of the shield.

o Approximately 98 percent of the thermal shield activity is from cobalt-60, and the remaining activity mostly from nickel-63. (Note: 1977 data)

o Approximately 67 percent of the graphite activity is from tritium and 27 percent from carbon-14. (Note: 1977 data)

o Plutonium-239/240 concentrations in the graphite stack average 2.6×10^3 pCi/g up to a maximum of 2.7×10^4 pCi/g.

Some other general results reported in this study are as follows:

o In-place dose rates of selected process tubes at the F and H Reactors
   average approximately 100 R/hr in the vicinity of the gunbarrel tips and
   drop down to 8 R/hr 15 feet into the process tubes from the nozzle.

o Based on sampling performed in 1967, cobalt-60 concentrations in the
   DR Reactor process tubes are the order of 2.5×10^7 pCi/g. Cobalt-60
   concentrations in a mild steel gunbarrel tip removed from C Reactor are
   approximately 1×10^8 pCi/g, and 2×10^9 pCi/g in a stainless steel
   gunbarrel tip.

A.3 PAST PROCESS TUBE AND GUNBARREL SAMPLING

Sampling of the reactor hardware (process tubes, gunbarrels, etc.) was not
performed as part of this program. Examples of typical dose rates of
activated reactor hardware removed as part of maintenance projects during
operating periods are: gunbarrels - 300 mR/hr to 4 R/hr at 3 feet; step-
plugs - 100 mR/hr to 3 R/hr at 4 feet; horizontal control rods - 1 to 5 R/hr
at 6 feet; and bundles of process tubing - 500 mR/hr to 5 R/hr at 6 feet.

A limited amount of sampling of the DR Reactor process tubes was performed in
March 1967, a little more than 2 years after reactor shutdown. Sections of
aluminum process tubes #3768, #1166, and #2568 were removed and analyzed for
radionuclides. The film was removed from the interior of the tubes for
separate analyses. All samples were taken 14 feet from the rear face van
stone flange. The process tube connections are made to the gunbarrrels by van
stone flanges which were intended to prevent water from leaking into the stack
and the gas atmosphere. A cross section of a process tube channel for DR
Reactor is given in Figure A-3. The fuel element charges started
approximately 8.5 feet from the end of the van stone flange.

Table A-1 lists the radioanalytical results for the process tube samples.

Table A-2 lists the same results as decay-corrected until March 1977, the
midpoint of the reactor core sampling.

TABLE A-1

RADIOANALYTICAL RESULTS OF PROCESS TUBE SAMPLES

Radionuclide	Half-Life (T 1/2)	Process Tube Sections pCi/g Aluminum		
		#3768	#1166	#2568
Tube				
^{54}Mn	314 day	2.4×10^7	2.4×10^7	3.8×10^8
^{60}Co	5.3 year	9.4×10^7	9.2×10^7	4.6×10^8
Film				
^{54}Mn	314 day	3.9×10^5	3.0×10^5	-
^{60}Co	5.3 year	3.0×10^6	2.7×10^6	-
^{65}Zn	245 day	8.8×10^6	5.1×10^6	-
^{152}Eu	13 year	2.8×10^5	4.0×10^5	-
^{154}Eu	8 year	5.0×10^5	2.8×10^5	-
^{90}Sr	28 year	2.0×10^4		-

TABLE A-2

RADIOANALYTICAL RESULTS OF PROCESS TUBE SAMPLES
DECAY - CORRECTED TO MARCH 1977

Radionuclide	No. of T 1/2's	Process Tube Sections pCi/g Aluminum		
		#3768	#1166	#2568
Tube				
^{54}Mn	12	5.9×10^3	5.9×10^3	9.3×10^4
^{60}Co	1.9	2.5×10^7	2.5×10^7	1.2×10^8
Film				
^{54}Mn	12	9.5×10^1	7.3×10^1	
^{60}Co	.1.9	8.0×10^5	7.2×10^5	
^{65}Zn	15	2.7×10^2	1.6×10^2	
^{152}Eu	0.77	1.6×10^5	2.3×10^5	
^{154}Eu	0.63	2.2×10^5	1.2×10^5	
^{90}Sr	0.36	1.6×10^4		

Figure A-3. Process Tube Channel Cross Section.

After 10 years of decay since the analyses were performed, cobalt-60 is the dominant radionuclide in the process tube wall. Cobalt-60, europium-152, europium-154, and strontium-90 are the primary radionuclides in the process tube film. Manganese-54 and zinc-65 have decayed to levels three orders of magnitude less than the other radionuclides. Manganese-54 and zinc-65 would probably be masked by the activity of the other radionuclides, if gamma analyses of process tube samples were now performed.

In April 1967 samples from the tips of two gunbarrels removed from C Reactor were analyzed. The cobalt-60 results for these two samples are given below.[1]

o   C Reactor Gunbarrel (4475) - 304 stainless steel

$$^{60}Co = 3.2 \times 10^9 \text{ pCi/g} \quad \text{(Redox Lab Analysis)}$$
$$^{60}Co = 7.9 \times 10^9 \text{ pCi/g} \quad \text{(Purex Lab Analysis)}$$

o   C Reactor Rear Gunbarrel (0954) - mild steel

$$^{60}Co = 4.4 \times 10^8 \text{ pCi/g} \quad \text{(Redox Lab Analysis)}$$

Decay-correcting these data to March 1977 gives an average concentration of 1.5×10^9 pCi/g for the stainless steel gunbarrel, and 1.2×10^8 pCi/g for the mild steel gunbarrel. It should be noted that normal gunbarrels are mild steel. The stainless steel gunbarrel was one of about six prototypes installed on C Reactor for testing.

Dose rate measurements using a shielded VCP (with window closed) were taken of the mild steel gunbarrel to determine the extent of the gunbarrel tip activation. Figure A-3 illustrates the dose rate measurements taken along the tip of the gunbarrel. The dose rate information indicates that about a third of the gunbarrel was activated. (See Figure A-4.) The biological and thermal

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[1]Other short half-life radionuclides (iron-59, cobalt-61, etc.) were detected, but would have since decayed away.

POSITION (INCHES)	DOSE RATE (mR/hr)
0	580
5	870
7	730
13	670
16	480
19	240
22	150
25	85

Figure A-4. Dose Rates to Determine Gunbarrel Tip Activation
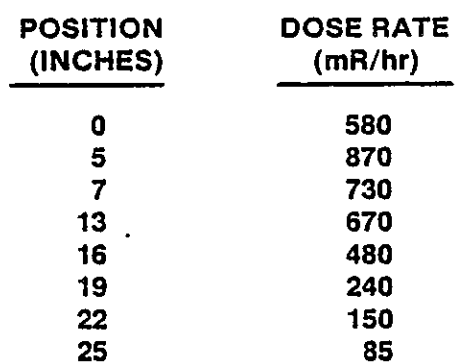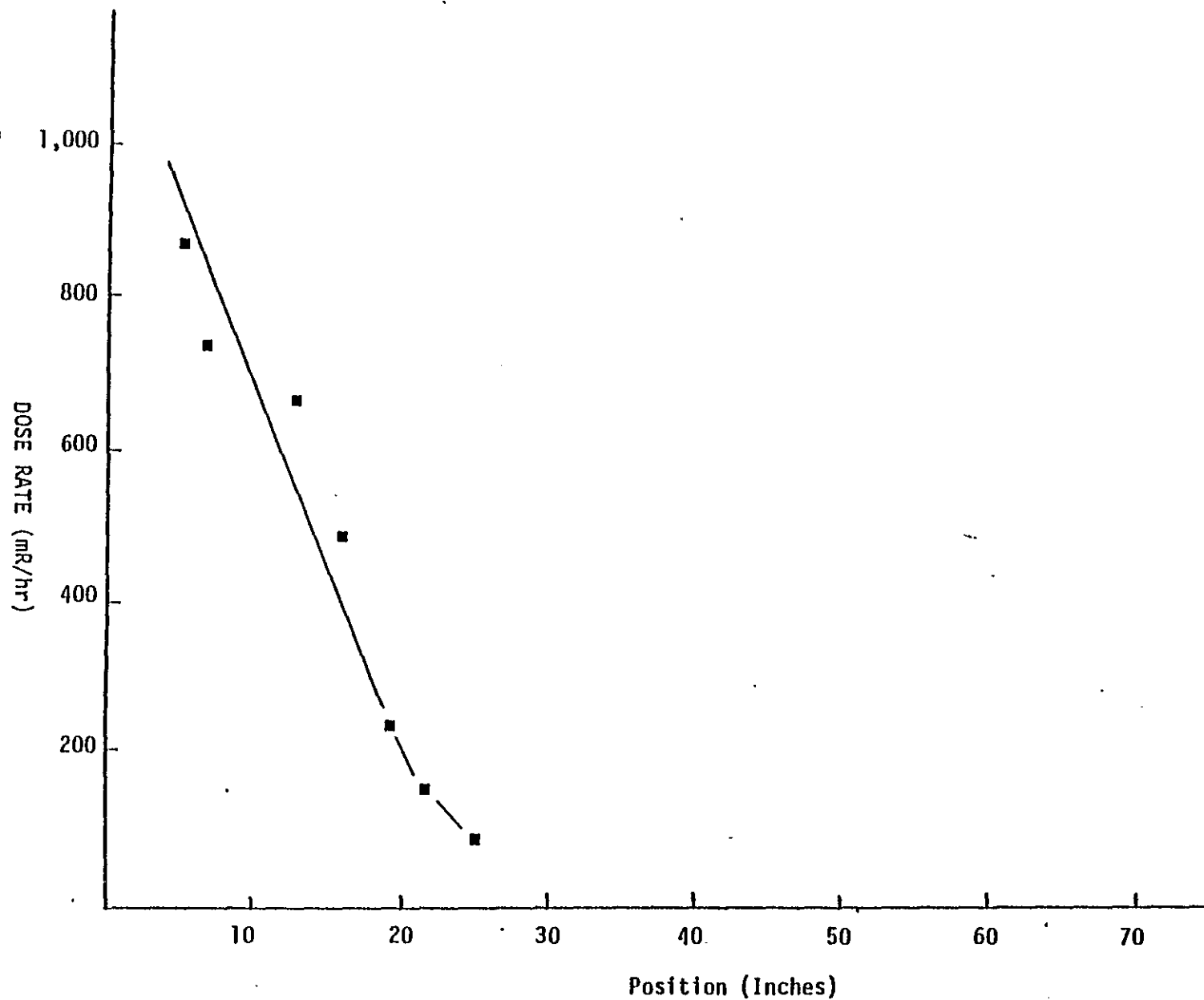
Figure A-5.   C Reactor Gunbarrel Dose Rate Measurements at 2 Feet vs. Detector Position Line Along Gunbarrel Tip.

shield sampling performed as part of this study also indicated that the inner third of a gunbarrel will be activated.

A.4   DISCUSSION OF THE DR REACTOR CORE SAMPLING AND RESULTS

The approximate locations of the four sample holes drilled into DR Reactor from the X-levels were given in Figure A-2.   A more precise description of the sample hole locations follows:

    Sample Hole A - Located on the X-2 level of the right side of the reactor.
       The hole is in graphite layer number 80 from the bottom of the graphite
       stack and is approximately 10 feet above the reactor horizontal
       centerline and 5 inches left of the vertical centerline.[2]

    Sample Hole B - Located on the X-2 level of the reactor.   The hole is in
       graphite layer number 82 and is approximately 10 feet-8 inches above the
       reactor horizontal centerline and 7 feet-10 inches to the left of the
       vertical centerline.

    Sample Hole C - Located on the X-1 level of the reactor.   The hole is in
       graphite layer number 58 and is about 2 feet-3 inches above the reactor
       horizontal centerline and 1 foot-2 inches left of the vertical
       centerline.

    Sample Hole D - Located on the X-0 level of the reactor.   The hole is
       2 feet-6 inches above the bottom row of process tubes and is between the
       4th and 5th rows of process tubes.   The hole is 13 feet below the
       reactor horizontal centerline and 7 feet right of the vertical
       centerline.

Basic sampling criteria were as follows:

o   Biological shield samples were drill cuttings of approximately 50 grams.
    The sampling interval of the biological shield samples depended upon the
    contamination levels encountered as measured by field instrumentation.

o   Thermal shield samples were 5/8-inch diameter core samples.   Small aliquots
    were then taken for radioanalytical analyses.

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[2]There are 102 graphite layers in the DR Reactor graphite stack.

o Graphite stack samples were 1/2-inch diameter core samples taken approximately every 5 feet. The sample size was about 20 grams of graphite.

A.4.1 Biological Shield Samples

Reactor core samples were numbered for location and material type. 105-DR represents the DR Reactor building number.

a) Masonite sample - 105-DR-AM1, where "A" designates the sample hole, and "M1" indicates that it was the first layer of masonite encountered in sample hole A. The second layer of masonite would be designated as 105-DR-AM2, etc.

b) Iron sample - 105-DR-AI1, where "A" designates the sample hole, and "I1" indicates that it was the first layer of iron encountered in sample hole A.

A.4.2 Thermal Shield Core Samples

A thermal shield core sample is numbered 105-DR-AT, where "A" designates the sample hole and "T" that the sample is a thermal shield core sample.

Thermal shield samples were taken as continuous core samples, which were later segmented into smaller samples. Small aliquotes were taken as drill shavings collected at quarter-inch intervals along the core samples with the exception of test hole A. For test hole A, the thermal shield core sample was cut into quarter inch thick disks. Drill shavings were then taken by drilling through the face of the quarter inch thick disks. Drill shaving samples have suffixes of the form T1, T2, etc., where T1 indicates the inner quarter inch sample, T2 the following quarter inch sample, etc.

A.4.3 Graphite Samples

A reactor graphite sample is numbered 105-DR-AG15, where "A" designates the sample hole, and "G15" that it is a graphite sample taken from 15 feet into the reactor core.

A.4.4 Sample Analysis and Quality Control

Sampling, documentation and analysis were performed in accordance with written procedures using trained, experienced personnel. Samples were analyzed for gamma emitters by UNC's radioanalytical laboratory. All other radionuclide analyses were performed by U.S. Testing, Richland, Washington. Quality control on sample analysis consisted of resubmitting a certain percentage of samples under a different sample number and verifying the consistency of results. Both labs had documented QA/QC controls in place and both participated in the EPA intercomparison program.

A.4.5 In-Place Measurements

In-place dose rates and Geiger-Mueller (GM) meter counts of the biological shield, iron-drill shaving samples for the four sample holes are shown in Figure A-6. The GM counts for the drill shaving samples are only partially representative of the specific activities because of possible variations in sample sizes. In-place dose rates and GM counts of the samples were highest for sample hole A, even though sample hole C is closest to the center of the reactor.

Radioactivity was not detected with field instrumentation in the iron samples of the biological shield until the inner fifth layer for sample holes A and C, the sixth layer for sample hole B, and the last layer for sample hole D. In-place dose rates for sample holes B and C were essentially the same, ranging from 0.5 mR/hr at the start of the fifth layer of iron up to 200 mR/hr at the start of the last layer of iron. In-place dose rates for sample hole A
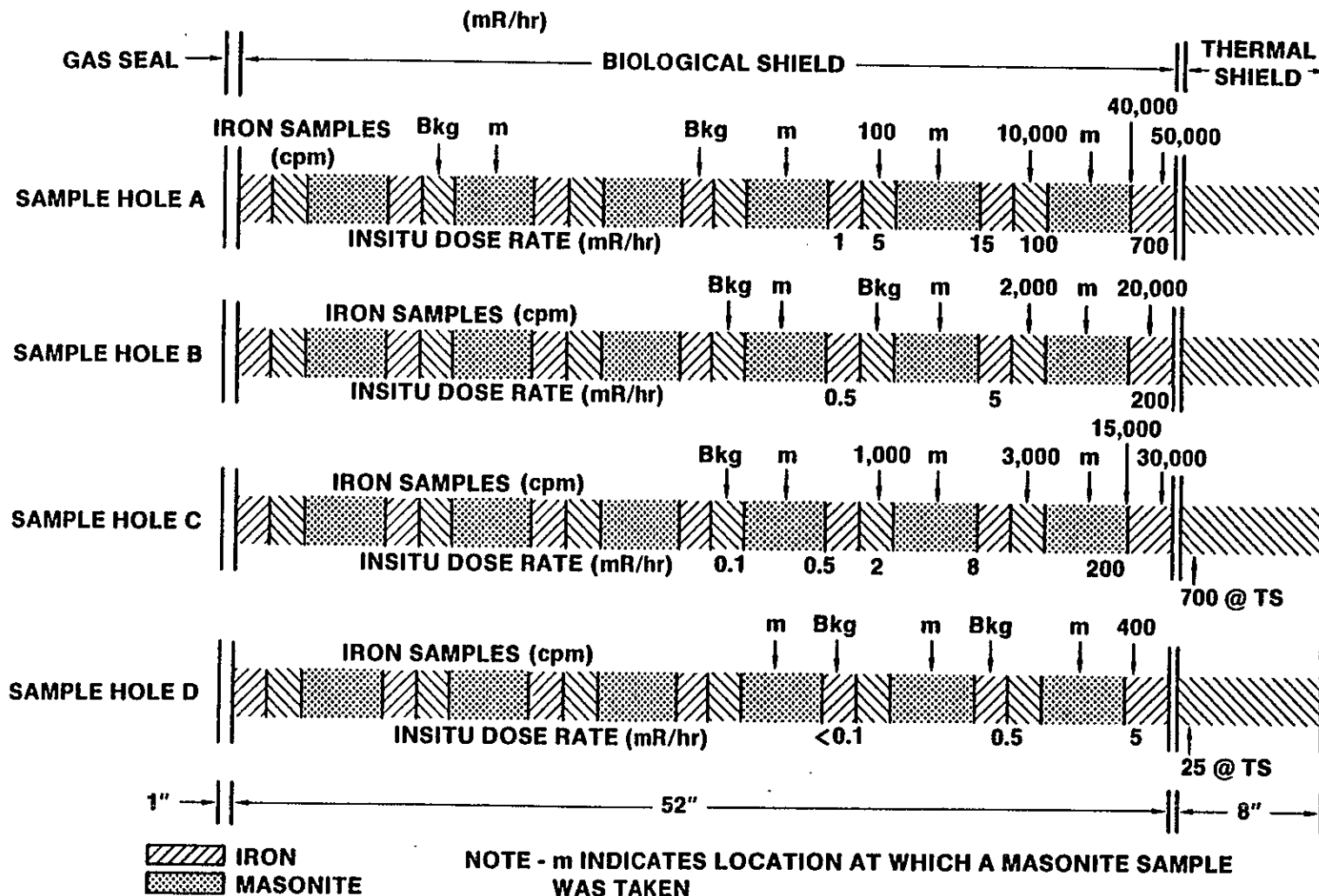
Figure A-6.   In-Place Dose Rates Within DR Reactor Sample Holes.

were the highest, ranging from 1 to 700 mR/hr for the same interval. Sample hole D had the lowest in-place dose rates which ranged from 0.1 mR/hr at the start of the fifth iron layer up to only 5 mR/hr at the start of the last layer of the biological shield.

No activity measurable with field instrumentation was detected in any of the Masonite samples, except for the last layer for sample hole A which read 1,000 cpm with a GM probe. This activity, however, is attributable to contamination from drill shavings which were visibly present in the sample. The contamination was removed from this Masonite sample by using a magnet.

Cobalt-60 concentrations for selected samples from the biological shields followed the same pattern indicated by field instrumentation, except that low concentrations of cobalt-60 were detected in the fourth layer of iron as shown in Table A-3.

TABLE A-3

COBALT-60 CONCENTRATION IN BIOLOGICAL SHIELD

Biological Shield Iron Layer	^{60}Co Concentration (pCi/g)			
	Hole A	Hole B	Hole C	Hole D
7	1.3×10^5	5.4×10^4	5.4×10^4	4.0×10^3
6	1.7×10^4	6.8×10^3	3.0×10^3	detect.
4	3.3×10^1	1.7×10^1	detect.	detect.

A comparison of thermal shield samples for these sample holes also shows the same trend (Table A-4). Sample holes B and C have similar concentrations, but hole A has concentrations about four times greater than B and C.

TABLE A-4

COBALT-60 CONCENTRATIONS IN THERMAL SHIELD

Distance from Inner Surface of Thermal Shield	^{60}Co Concentration (pCi/g)			
	Hole A[3]	Hole B	Hole C	Hole D
1/4"	4.0×10^8	7.4×10^7	7.6×10^7	5.0×10^7
1/2"	1.8×10^8	4.8×10^7	4.6×10^7	3.5×10^7
1-1/2"	2.6×10^7	9.0×10^6	1.8×10^7	6.4×10^6
3"	4.7×10^6	1.4×10^6	3.1×10^6	9.6×10^5

More complete summaries of the radionuclide analyses of selected biological and thermal shield samples are included in Tables A-5 and A-6, respectively. The data indicate that approximately 90 percent of the total thermal and biological shield cobalt-60 activity is within the inner 1-3/4 inch of the thermal shield, and 99 percent of the activity is in the inner 4 inches of the thermal shield. The cobalt-60 activity within the inner half of the thermal shield has an average concentration of 3.6×10^7 pCi/g.

Nickel-63 is also present in the thermal and biological shields. Because of the high expense of performing nickel-63 analyses, not all samples were analyzed for nickel-63. The highest nickel-63 concentration found within the inner 4 inches of the thermal shield was 7.5×10^6 pCi/g.

Graphite samples had direct GM counts generally about 10,000 cpm, but ranged from 1,000 cpm to over 80,000 cpm. Uncorrected dose rate measurements of the samples with a Cutie Pie (CP)[4] were generally 1 mR/hr up to a maximum of 20 mR/hr for sample A-G33.

---

[3]Sample hole A results are adjusted for the difference in the technique used for segmenting the thermal shield core sample for this test hole; i.e., the concentration at a quarter inch is the average of samples T-1 and T-2, etc.

[4]Cutie Pie is the name given a hand-held, ionization-chamber-type, radiation detection instrument.

TABLE A-5

DR REACTOR BIOLOGICAL SHIELD IRON SAMPLES

Concentration (pCi/g)

Sample No.	^{60}Co	^{63}Ni
A-I2	2.6×10^{-1}	
I4	3.3×10^{1}	
I5	3.7×10^{2}	1.2×10^{1}
I6	1.7×10^{4}	
I7	1.3×10^{5}	3.4×10^{3}
I7-1/2	9.7×10^{4}	
(3/4" core) I8	2.5×10^{5}	
B-I4	1.7×10^{1}	5.7×10^{-1}
I6	6.8×10^{3}	
I7	5.4×10^{4}	8.2×10^{2}
C-I4	*	
I5	2.5×10^{3}	4.1×10^{1}
I6	3.0×10^{3}	
I7	5.4×10^{4}	
I7-1/2	6.8×10^{4}	7.8×10^{2}
D-I4,5,&6	*	
I7	4.0×10^{3}	6.6×10^{1}

*Less than analytical detection limit. Detection limit value data no longer available.

TABLE A-6

DR REACTOR THERMAL SHIELD SAMPLES

Concentration (pCi/g)

	Sample No.*	^{60}Co	^{63}Ni		Sample No.	^{60}Co	^{63}Ni
	A-T1	5.4×10^8	4.8×10^6		C-T1	7.6×10^7	7.5×10^6
	T2	2.6×10^8		(1/2")	T2	4.6×10^7	
	T3	9.1×10^7		(1-1/2")	T6	1.8×10^7	
(1")	T4	7.8×10^7		(3")	T12	3.1×10^6	
	T5	4.6×10^7			T18	8.2×10^5	
	T6	2.4×10^7		(6")	T24	2.5×10^5	6.9×10^3
	T7	2.7×10^7		(7-1/4")	T26	1.4×10^4	
(2")	T8	2.5×10^7					
	T9	2.7×10^7			D-T1**	5.0×10^7	9.2×10^5
	T10	1.4×10^7			T2	3.5×10^7	
	T11	8.8×10^6			T3	2.1×10^7	
(3")	T12	5.2×10^6		(1")	T4	1.2×10^7	
	T13	4.1×10^6			T5	9.2×10^6	
	T14	8.6×10^6			T6	6.4×10^6	
	T15	2.5×10^6			T7	4.2×10^6	
(4")	T16	2.6×10^6	3.9×10^4	(2")	T8	4.2×10^6	
	T17	1.6×10^6		(3")	T9	9.6×10^5	
				(4")	T10	2.4×10^5	
	B-T1	7.4×10^7	2.9×10^6	(5")	T11	7.8×10^4	1.3×10^3
	T2	4.8×10^7					
	T6	9.0×10^6					
(3")	T12	1.4×10^6					
	T18	3.2×10^5					
(5")	T20	2.3×10^5	5.0×10^3				

*Thermal shield samples were taken as continuous 5/8-inch diameter cores. Drill shaving samples were taken at quarter-inch intervals and designated as T-1, T-2, etc.

**For sample hole D, drill shaving samples were taken at 1/4-inch intervals for the first 2 inches, and then at 1-inch intervals.

Radionuclide analyses for selected graphite samples from the DR Reactor are given in Table A-7. Tritium and carbon-14 are present in addition to mixed fission and activation products. Transuranics are also present.

Plutonium-239/240 concentrations average 2.6×10^3 pCi/g. Sample A-G33 had a plutonium-239/240 concentration of 2.7×10^4 pCi/g. Nickel-63 was present with concentrations of up to 1.5×10^5 pCi/g.

At DR Reactor, there was enough stack separation and graphite breakage so that balls were known to have been retained in the stack structure after spurious ball-drops. In fact, one ball was recovered in a graphite sample taken from sample hole A drilled into the DR Reactor stack. The ball had uncorrected CP readings of 10 R/hr at one inch and 150 mR/hr at one foot. This undoubtedly was the case to various degrees at the other reactors as well. The ball 3X systems of the 100 Area production reactors utilized both 3/8-inch and 7/16-inch diameter nickel-plated boron steel, nickel-plated carbon steel, and stainless steel balls which dropped into the vertical safety rod channels after a trip of the ball 3X circuit. The nickel plating and nickel impurities in the stainless steel would be sources of nickel-63 radioactivity. Balls retained in the graphite stack would have been subjected continuously during reactor operation to the neutron flux, heat, and moisture within the stack.

All analytical results are accurate to two standard deviations. Additionally, for gamma-emitting radionuclides, the error is no greater than 30 percent of the result. Results with a greater error were recounted until the error was within the 30 percent limit.

A.5 REACTOR IN-PLACE DOSE RATE MEASUREMENTS

During February 1978 in-place dose rate measurements were taken in selected process tubes at the DR and F Reactors. The in-place dose rate measurements were taken using a Tele-probe. Similar measurements were taken at the H Reactor in August 1975. Readings were taken from both the front and rear

TABLE A-7

DR REACTOR GRAPHITE SAMPLES

Concentration (pCi/g)

Sample Number	Pu-238	Pu-239/240	Sr-90	U	^{3}H	Eu-152	Co-60	Eu-154	Cs-134	Cs-137	Eu-155	C-14	Ba-133	Ni-63
A-G 6						5.6×10^4	8.0×10^4	4.6×10^4	1.0×10^3	2.0×10^3	1.2×10^4	5.4×10^5	4.9×10^3	1.2×10^5
G 7						4.3×10^4	6.1×10^4	1.7×10^4	3.2×10^3	1.1×10^3	4.5×10^3		3.5×10^3	
G 13	1.3×10^2	2.0×10^2	3.5×10^3	6.4×10^{-1}	9.8×10^6	7.7×10^4	1.1×10^5	1.8×10^3	1.1×10^3	7.1×10^3	1.1×10^3	2.7×10^6	7.4×10^3	
G 18	$<1.4 \times 10^2$	$<3.8 \times 10^1$	1.7×10^3	$<2.0 \times 10^0$	1.3×10^7	1.9×10^3	2.7×10^3	9.9×10^1	1.2×10^2	3.0×10^3	1.3×10^2	7.1×10^6	3.4×10^3	
G 23						3.4×10^4	4.8×10^4	7.8×10^3	2.5×10^3	2.1×10^3	2.2×10^3	1.2×10^7	2.4×10^3	
G 28	$<7.8 \times 10^1$	3.9×10^2	5.6×10^3	$<1.1 \times 10^0$	7.1×10^6	1.3×10^5	1.8×10^5	6.6×10^3	1.8×10^3	1.6×10^4	1.9×10^3	4.3×10^6	1.4×10^4	
G 33*	4.7×10^4	2.7×10^4	7.5×10^6	8.1×10^{-2}	6.3×10^6	1.1×10^4	1.2×10^5	2.1×10^4	2.2×10^4	5.7×10^5	5.8×10^3	5.0×10^6	3.4×10^4	
B-G 6						5.7×10^2	8.2×10^2	1.4×10^2	2.1×10^1	3.5×10^2	3.8×10^1	8.2×10^5	2.8×10^2	
G 13	7.8×10^0	2.7×10^1	1.0×10^3	1.9×10^0	4.7×10^6	4.8×10^2	6.9×10^2	1.2×10^2	3.3×10^1	1.6×10^2	3.2×10^1	1.8×10^6	1.3×10^2	
G 18						6.2×10^3	8.9×10^4	1.0×10^4	1.4×10^3	1.3×10^4	2.7×10^3		1.0×10^4	
G 23	7.7×10^1	1.6×10^2	2.8×10^3	$<6.2 \times 10^{-2}$	7.8×10^6	1.3×10^5	1.8×10^5	3.8×10^4	3.2×10^3	1.1×10^4	1.1×10^4	2.2×10^6	1.8×10^4	
G 28						2.1×10^4	3.0×10^4	5.9×10^3	2.6×10^3	1.1×10^4	1.1×10^3		4.4×10^3	
G 33	2.0×10^2	4.9×10^2	3.8×10^3		3.4×10^6	3.6×10^4	5.1×10^4	5.1×10^3	1.7×10^3	9.3×10^3	1.3×10^3	1.8×10^6	2.6×10^3	
C-G 7	2.9×10^1	$<1.2 \times 10^{-1}$	1.0×10^3	$<1.3 \times 10^{-1}$	2.4×10^6	1.2×10^5	4.3×10^4	1.4×10^5	9.7×10^2	1.6×10^3	2.9×10^4	3.8×10^5	5.5×10^3	5.1×10^4
G 13						3.3×10^2	8.9×10^2	3.4×10^2	2.0×10^1	4.5×10^1	7.6×10^1	2.8×10^6	7.7×10^1	
G 16	$<1.0 \times 10^1$	$<9.0 \times 10^0$	1.4×10^3	$<1.0 \times 10^0$	4.1×10^6	4.9×10^1	1.3×10^2	5.0×10^1	2.9×10^0	1.3×10^3	1.1×10^1	2.2×10^5	2.2×10^3	
G 23	4.9×10^3	5.1×10^3	5.0×10^4	$<2.5 \times 10^0$	5.0×10^6	1.9×10^3	8.5×10^3	3.3×10^3	1.7×10^3	6.5×10^4	7.2×10^2	3.3×10^6	1.1×10^5	1.5×10^4
G 28						7.9×10^2	2.1×10^3	8.1×10^2	4.7×10^1	2.8×10^3	1.8×10^2	1.5×10^6	4.8×10^3	
G 33						1.4×10^4	3.1×10^5	1.6×10^4	5.4×10^3	2.4×10^4	4.2×10^3	1.5×10^6	4.8×10^4	
G 38	2.5×10^2	2.0×10^2	5.3×10^3	$<1.8 \times 10^0$	4.2×10^6	9.4×10^4	2.5×10^5	7.4×10^4	5.6×10^3	1.5×10^4	1.8×10^4	1.6×10^6	1.9×10^4	
D-G 7						2.4×10^5	4.5×10^4	2.8×10^5	7.1×10^2	3.0×10^3	4.5×10^4	6.1×10^5	8.2×10^3	
G 13	7.0×10^1	1.8×10^2	2.6×10^3	$<2.6 \times 10^{-1}$	1.3×10^7	4.4×10^4	8.6×10^4	4.5×10^4	1.4×10^3	4.7×10^3	1.1×10^4	1.6×10^6	1.7×10^4	
G 18						1.6×10^4	6.0×10^4	1.9×10^4	2.6×10^3	1.0×10^4	4.0×10^3	2.7×10^6	1.3×10^3	
G 23	2.7×10^2	2.4×10^2	2.7×10^3	$<4.0 \times 10^0$	7.7×10^6	1.0×10^5	2.0×10^5	1.0×10^5	3.2×10^3	1.3×10^4	2.4×10^4	2.0×10^6	1.7×10^4	1.5×10^5
G 28						1.8×10^5	1.8×10^5	1.8×10^5	1.2×10^3	1.0×10^4	7.4×10^3		1.9×10^4	
G 33						1.1×10^5	1.1×10^5	8.8×10^4	1.7×10^3	5.7×10^3	1.8×10^4		1.4×10^4	

*Am-241 3.8×10^3
Blanks in the table indicate that no analysis for that radionuclide was performed on a particular sample.

faces up to 15 feet into the process tube from the end of the nozzle. Fifteen feet was the greatest distance the Tele-probe could be extended. The results of these readings are given in Table A-8. Two measurements are generally reported for each of the process tubes measured. The first reading is the maximum dose rate detected at the end of the gunbarrel, or about 9.5 feet from the nozzle. In two cases, three readings are reported. In these instances, in-place dose rate readings went down going away from the gunbarrel tip and then went back up, thereby indicating higher than normal activity in the graphite stack.

As part of the 100-F Area Decontamination & Decommissioning (D&D) program, additional in-place dose rate surveys of process tubes were initiated and completed on June 11, 1979. The instrument used was ZETEX-302A with a 40-foot extension cable.

Readings of 37 selected process tubes were taken at predetermined points. The highest readings were found in the area of the front thermal shield and the front gunbarrel tips. The maximum reading was 251 R/hr with the average being 120 R/hr. Seven localized hot spots were found reading 15 to 78 R/hr; they were located in a random pattern but within 5 feet of the reactor center. The general background in this area inside the reactor is approximately 5 to 10 R/hr.

TABLE A-8

PROCESS TUBE IN-PLACE DOSE RATE MEASUREMENTS

Reactor	Process Tube No.	Dose Rate at Gunbarrel Tip (R/hr)	Dose Rate 15 ft in from Nozzle (R/hr)
DR Reactor Front Face	0459	28	2.7
	0472	43	3
	0488	35	3.5
	1476	50	Not measured
	2361	70	Not measured
	2374	80	5
	2386	50	Not measured
DR Reactor Rear Face	0659	50	
	0673	70	
	0690	40	
F Reactor Front Face	0459	65	2.5
	0473	75	4
	0488	110	7-24*
	2173	150	4-6 *
F Reactor Rear Face	0659	30	Not measured
	0673	45	Not measured
	0688	27	Not measured .
H Reactor Front Face	1873	300	12
	0673	100	8
	0473	65	8

*In-place dose rate readings went down going away from the gunbarrel tip (1st No.) and then went back up (2nd No.).

APPENDIX B

GRAPHITE SAMPLING OF KW REACTOR

1. Graphite Sample Locations

2. Letter, J. C. Sheppard to J. Stevenson, "C-14 Analysis of Graphite Samples," dated July 24, 1984

3. Letter, W. C. Morgan to J. A. Adams, "Results of Carbon-14 Measurements," dated June 28, 1984

4. Discussion of Sample Analyses

5. Carbon-13 Activation

6. Letter, W. C. Morgan to J. A. Adams, "Results of Nickel-63 Analysis," dated June 7, 1985

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REACTOR GRAPHITE STACK

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21

Channel #1880

19
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11
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7
5
3
1

41 43   45 47   49   5, 53   55 57   59   61 63   65 67   69   71   73   75 77   79   81 83   85 87   89   91   93   95 97 98

H O R I Z O N T A L   T U B E   R O W S   41 - 48

GRAPHITE SAMPLE LOCATION
KW REACTOR BLOCK
(FRONT VIEW)
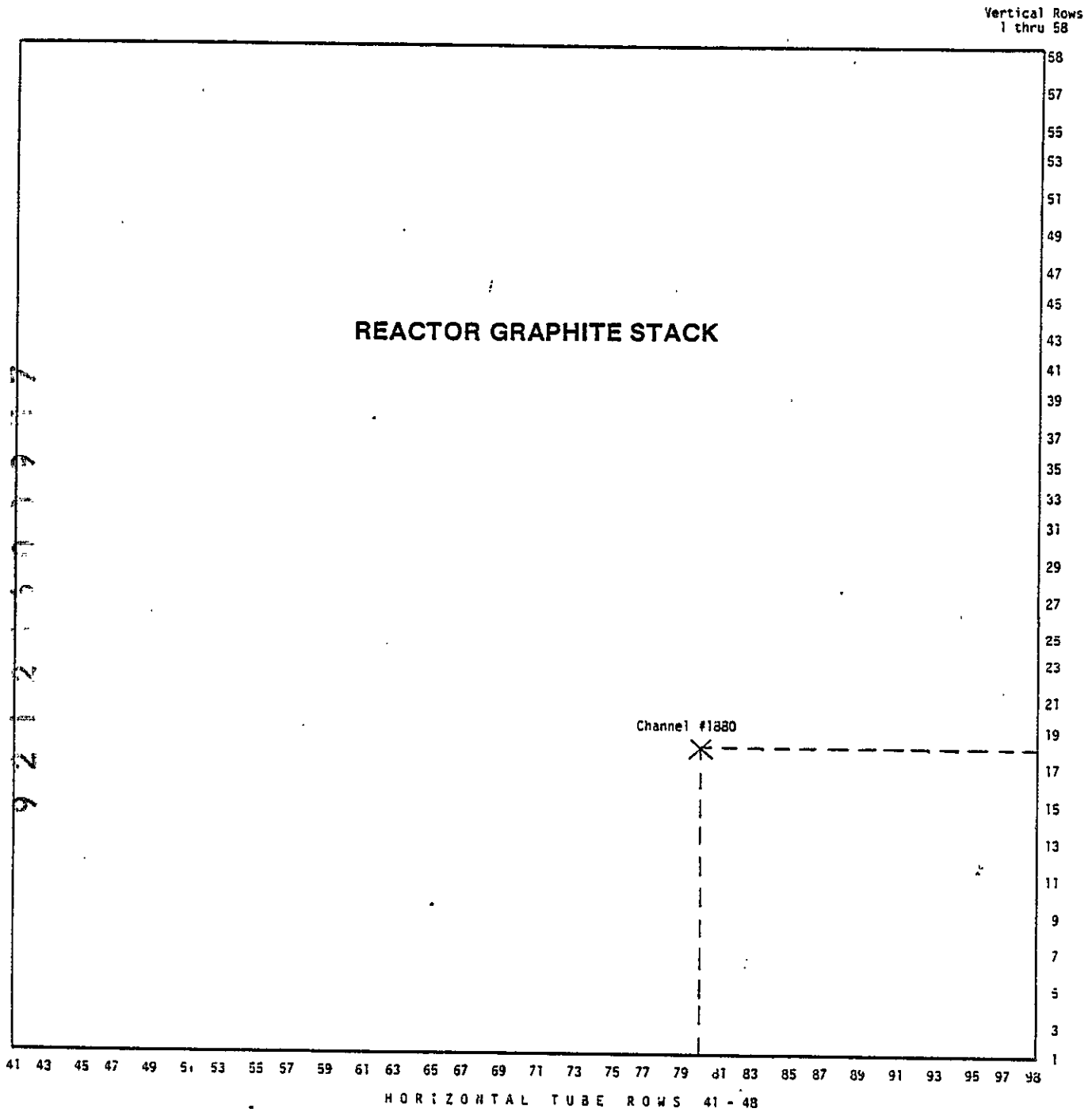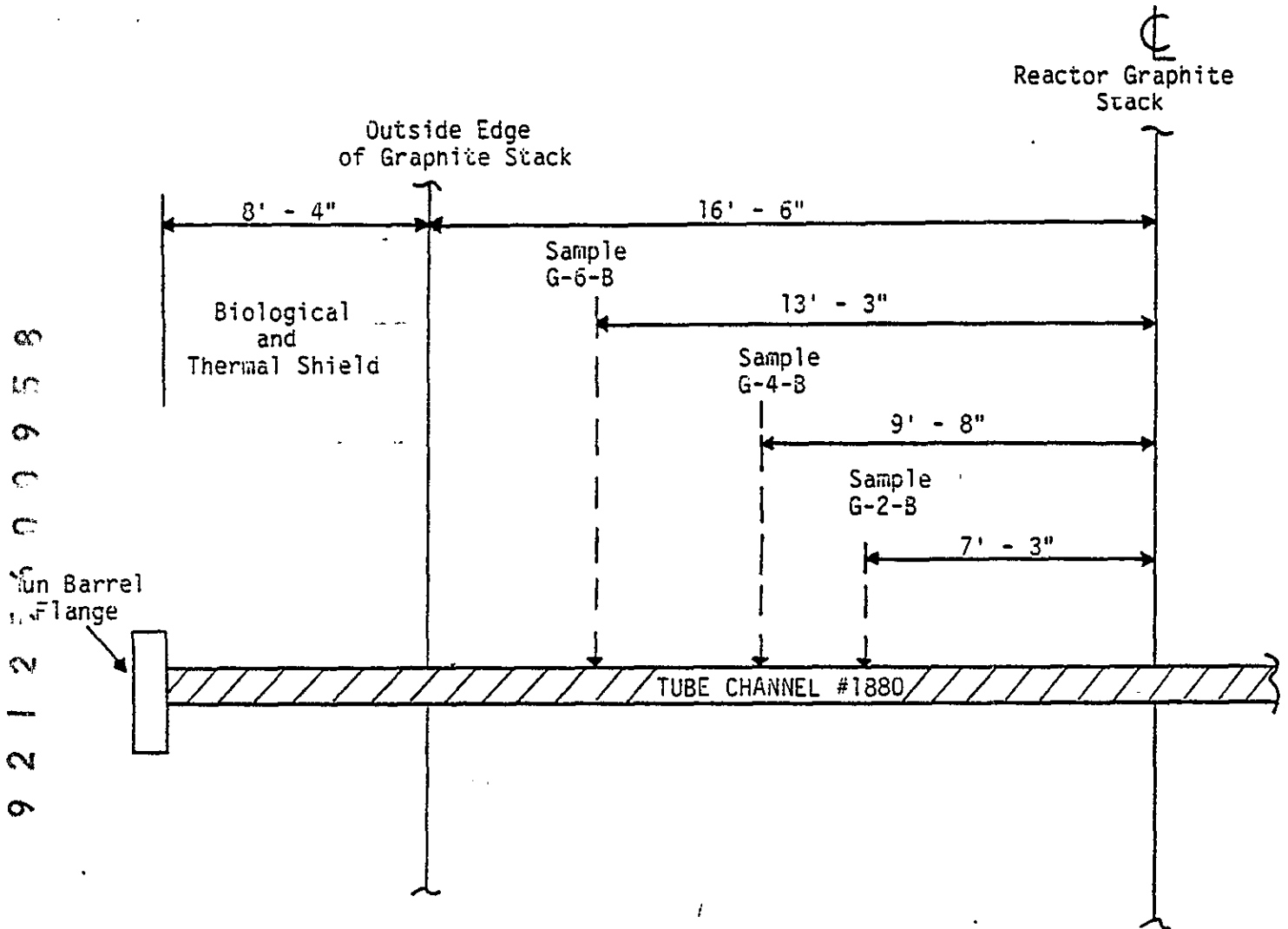
**GRAPHITE SAMPLING LOCATIONS
KW REACTOR BLOCK
(SIDE VIEW)**

Washington
State University

Department of Chemical Engineering, Pullman, Washington 99164-2710 / 509-335-4332

July 24, 1984

Mr. Jim Stevenson
UNC Nuclear Industries
P.O. Box 490
Rm 430, Federal Building
Richland, WA 99352

Dear Mr. Stevenson:

Here is our report on the ^{14}C analysis of your graphite samples.

All samples were burned in oxygen at $650^{\circ}C$ to form CO_2^-. Known volumes of CO_2 were injected in a 450 cm^3 gas proportional counting tube with high purity methane and counted until 1000 to 2000 counts were accumulated. Thus, the counting error ranged between 2.2 and 3.2 percent. Backgrounds of about 2.5 cpm were subtracted off, even though they were obviously negligible.

National Bureau of Standards oxalic acid, the radiocarbon dating standard, was used to obtain the detection efficiency of this detector. At $25^{\circ}C$ and 2.00 atmospheres methane, the detector contains 0.442 grams of carbon with a specific activity of 13.6 disintegration per minute per gram of carbon (the equivalent of the NBS oxalic acid). Thus, the disintegration rate should be 0.442 x 13.6 or 6.01 dpm. The observed, long-term average, counting rate for this detector is 5.10 ± 0.01 counts per minute, therefore our detector efficiency is 84.8 percent very close to the value found for similar detectors. In all counts, this efficiency correction was made. With the exception of one sample, all values represent an average of two or more separate counts, sometimes with significantly different sample sizes.

The Table following summarizes our results.

WSU No.	UNC I.D. No's	$cpm/cm^3 CO_2$	dpm/g of carbon
3003	105-DR, DG-33-3	2460	5.91×10^6 (a,b,c)
3004	" , BG-18-3	2227	5.35×10^6
3005	" , BG-28-3	959	2.30×10^6
3006	" , DG-28-3	880	2.11×10^6
3007	" , AG-7-3	360	8.64×10^5
3016	C-3 Inert	<1	$<<2.4 \times 10^3$
3022	105-KW, G-4-B	1327	3.10×10^6
3023	" , G-6-B	747	1.79×10^6
3024	" , G-2-B	3396	8.15×10^6

(a) $\text{dpm/g of carbon} = cpm/cm^3 \times 22{,}400 \frac{cm^3}{mole} \times \frac{1 \text{ mole}}{12.01 \text{ g carbon}} \times \frac{1}{0.848}$

(b) Estimated error somewhere between 5 to 10 percent.

(c) Divide by 2.22×10^6 to specific activity in microCuries per gram of carbon.

As mentioned above, the estimated errors are in the 5 to 10 percent range. The counting errors, an uncertainty in the counting tube volume and the so-called end-effect contribute to this error. It would take a much more intense research program to obtain higher precision results.

We had expected lower ^{14}C specific activities, however my calculations suggest that the specific activities reported above may not be too far off. Assuming the following:

$$\sigma_{13} = 0.9 \times 10^{-27} cm^2 \quad ^{13}C \text{ abund} = 0.0111$$

$$\lambda_{14} = 0.693/5740 \text{ yr}^{-1}$$

$$\phi = 2 \text{ to } 3 \times 10^{13} \text{ n/cm}^2 \text{ sec}$$

$$\text{Irrad time} = 10 \text{ to } 30 \text{ years} = t$$
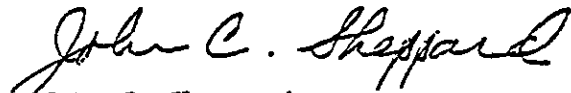
$$A_{14} = N_{13} \sigma_{13} \phi \lambda_{14} t \quad dps$$

For 10 years at a flux of 2×10^{13}, we obtain a specific activity of 7.27×10^5 dpm/g of carbon.

For 30 years at a flux of 3×10^{13}, the calculated specific activity is 3.27×10^6 dpm/gram of carbon. Our calculations are in general agreement with the measurements.

If you have questions, please phone us.

Sincerely,

John C. Sheppard
Professor of Chemical Engineering
 and Anthropology

JCS/kw

Battelle
Pacific Northwest Laboratories

Date   June 28, 1984

To   J. A. Adams

From   W. C. Morgan

RECEIVED
JUL 03 1984
J.A. ADAMS

PO Jackson
WE Kennedy
CW Thomas
LB/File

Subject Results of Carbon-14 Measurements

Our Radiological and Inorganic Chemistry Section obtained the following results for carbon-14 activities from the graphite samples submitted to them.

Reactor	Sample No.	^{14}C Activity, $\mu Ci/g$ $(\pm 2\sigma)$*
--	Control	$<3.25 \times 10^{-4}$
DR	A-G7-2	0.67 ± 0.02
DR	B-G18-2	2.36 ± 0.04
DR	D-G28-2	2.34 ± 0.03
DR	D-G33-2	1.71 ± 0.06
KW	G-2A	3.59 ± 0.01
KW	G-4A	2.18 ± 0.02
KW	G-6A	1.62 ± 0.02

Measurement of the ^{63}Ni activities on samples D-G33-2 and G-A4, should be finished within the next week or two.

_____

*   The error bar limits are counting statistics only, at twice the standard deviation.

WCM/tf

Discussion of Sample Analyses

The data provided by two independent laboratories, Washington State University (WSU) and Battelle Pacific Northwest Laboratories (PNL), show that the amount of carbon-14 measured varied by sample location and to some extent by the laboratory performing the analysis. For example, if the KW Reactor sample analyses are simply averaged and applied to the entire active region of the graphite stack, values of the carbon-14 are 6400 curies and 5100 curies, respectively for the PNL and WSU analysis values.

Another way to apply the sample analysis data is to assign the values to specific volumes of the reactor. For want of a better way to assign a volume to each sample, the graphite grade (see Appendix C) was used to define the volumes. The total volume of a K Reactor stack is 1.6×10^9 cm^3 (33.5' x 41' x 41'). The fraction of the stack represented by each grade zone (from Appendix C) are 40%, 30.5%, and 29.5% for the "green", "blue", and "red" zones respectively. Applying the G-2 sample value to the green zone, the G-4 sample value to the blue zone, and the G-6 sample value to the red zone, a carbon-14 value of 6680 curies is found from the PNL analyses. Similarly, a value of 5520 curies of carbon-14 is found from the WSU analysis.

To be conservative, a value of 6700 curies of carbon-14 was assumed for the KW Reactor.

Carbon-13 Activation

Carbon-14 originates from both the nitrogen-14 (n, p) carbon-14 reaction and the carbon-13 (n, gamma) carbon-14 reaction. A simplified calculation of the amount produced by the carbon-13 (n, gamma) carbon-14 reaction was performed. The activity of carbon-14 from the activation of carbon-13 can be represented by the expression:

$$A_{C-14} = f_2 N_0 \lambda / 3.7 \times 10^{10} \text{ curies}$$

where:

A_{C-14} is the carbon-14 activity per gram of graphite,

N_0 is the original number of atoms of carbon-13 per gram of graphite,

λ is the mean life of carbon-14,

f_2 is the product, t, where is the capture cross-section of carbon-13, is the thermal flux, and t is the irradiation time.

Substituting in the values for the parameters of the equation and assuming a flux of 10^{14} neutrons/cm^2-s for a time t of 15 years we have:

$$\lambda = \frac{.693}{\text{half-life of C-14}} = \frac{.693}{5730 \text{ years}} \doteq 3.84 \times 10^{-12} \text{ sec}^{-1}$$

$$N_0 = \frac{(.0111)(6.02 \times 10^{23})}{12.01} = 5.56 \times 10^{20} \text{ atoms C-13/gram}$$

$$\sigma = 1.4 \times 10^{-27} \text{ cm}^2$$
$$\phi = 10^{14}/\text{cm}^2\text{-s}$$

$$t = 15 \text{ yrs} \doteq 4.73 \times 10^8 \text{ seconds}$$

or

$$A_{C-14} = \frac{f_2 N_0}{3.7 \times 10^{10}} = \frac{(1.4 \times 10^{-27})(10^{14})(4.73 \times 10^8)(5.56 \times 10^{20})(3.84 \times 10^{-12})}{3.7 \times 10^{10}}$$

or

$$A_{C-14} = 3.83 \times 10^{-6} \text{ curies per gram of graphite.}$$

Within the active zone of a K reactor are about 1.6×10^9 grams of graphite, so for the total carbon-14 produced by the (n, gamma) reaction of Carbon-13 we have:

Active zone curies = $(3.83 \times 10^{-6})(1.6 \times 10^{9})$ = 6128 curies per reactor

A similar computation for the reflector region gives about 120 curies of carbon-14 per reactor. The combined active region plus the reflector region give a total value of about 6250 curies of carbon-14.

Date   June 7, 1985

To   J. A. Adams

From   W. C. Morgan

Subject Results of Nickel-63 Analysis

Our Radiological and Inorganic Chemistry Section obtained the following results for nickel-63 activities from the graphite core samples. Unfortunately, sample B-G18-2 from DR Reactor was somehow substituted for sample G-A4 from KW Reactor.

Reactor	Sample No.	nCi/g (±2σ)[*]
DR	B-G18-2	28.6 ± 1.2
DR	D-G33-2	50.9 ± 1.6

I am also enclosing a copy of a memo from C. W. Thomas, outlining the procedures used to determine the radionuclide activities in the graphite samples.

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[*] The error bar limits are counting statistics only, at twice the standard deviation.

ias

Enclosure

☀Battelle

Pacific Northwest Laboratories

Date   August 27, 1984   File/LB

To   W. C. Morgan

From   C. W. Thomas

Subject

Enclosed in the attached tables are the procedures used in identify-
ing the radionuclide and their concentrations in graphite samples
you recently sent us.

Table 1 is the procedure used in determining the radionuclides
present and some of their characteristics. Gamma spectrometric
measurements showed the samples to consist mainly of ^{60}Co, ^{154}Eu,
^{137}Cs, and ^{133}Ba. Recount after ignition showed the same isotopes
with insignificant reduction in concentrations. Beta absorption
studies showed essentially a single beta energy of 0.15 MeV. After
ignition less than 10% of the beta activity remained.

Table 2 shows the procedure used to determine the radionuclides
in the graphite samples that had a beta energy of 0.15 MeV. The
results from this procedure confirmed the beta activity was essen-
tially all ^{14}C.

Table 3 shows the procedure used to separate ^{63}Ni. The leaching
efficiency for removing ^{63}Ni from the graphite samples was deter-
mined by measuring the leaching efficiency of ^{60}Co. These data
showed the leaching efficiency was 81.0% for sample B-18 and 89.9%
for D-33. The radiochemical yield for recovery for ^{63}Ni was
determined by tracing with activated ^{65}Ni. The nickel yield for
B-18 was 86.5% while the yield for D-33 was 75.5%. The beta absorp-
tion studies confirmed the ^{63}Ni activity.

If you have any questions concerning the data and/or the procedures
used, don't hesitate to call.

CWT:dlc

Encl.

TABLE 1

Gamma and Beta Particle Characteristics Procedure.

An aliquot of sample was weighed and placed on a stainless steel dish. The gamma spectro was obtained by measuring on an intrinsic germanium detector. A beta energy analysis was obtained by counting this sample in thin window gas flow beta proportional counter using various thickness aluminum absorbers. The sample was then ignited using a butane burner. The residual material was again gamma and beta counted.

TABLE 2

Carbon-14 Procedure

Sample aliquot was placed in stainless steel boat with a small amount of copper oxide and inserted into a quartz tube which was placed in a tube furnace. A stream of oxygen was passed over the sample and the gaseous effluent was bubbled through a barium hydroxide trap. The tube furnace was slowly raised to $\sim$500 centigrade.

The barium carbonate from the trap was filtered and measured for ^{14}C.

TABLE 3

Nickel-63 Procedure

An aliquot of sample was weighed and gamma counted for ^{60}Co. The sample was then placed in hot 6M HCl, containing carriers and finely powdered by crushing with a glass rod. The sample was leached 3 times. The residue filtered and gamma counted. The leachates were combined and ^{65}Ni was added. The nickel was separated using dimethylglyoxime with citric acid complexing. The nickel was electroplated on a stainless steel disc and the ^{65}Ni measured for chemical yield in a gamma spectrometer and the ^{63}Ni measured in a windowless gas flow proportional beta counted AC shielded with NaI(Tl) detectors. Nickel-63 was verified by beta absorption measurements.

APPENDIX C

CHLORINE-36

Letter, W. C. Morgan to J. A. Adams, "Measured Versus Estimated Chlorine-36 Activities in Irradiated Graphite," October 30, 1986

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Battelle
Pacific Northwest Laboratories

Date   October 30, 1986

To   J. A. Adams

From   W. C. Morgan

Subject   <u>Measured Versus Estimated Chlorine-36 Activities</u>.
            <u>in Irradiated Graphite</u>

Conclusions

The measured activities of Cl^{36} in samples of graphite from KW Reactor are a
factor of 11.6 less than the estimated activities, which were based on the (con-
servative) assumption that the graphite retained all of its original chlorine
content. The reduction in Cl^{36} concentration was found to be independent of
neutron fluence. It is postulated that the residual Cl^{36} is due to chlorine
which became trapped in closed pores during initial reactor operation, while
most of the chlorine diffused out of the pores, which remained open. Other
data are sited in support of the above mechanism, which should apply equally
to all halogen purified graphites; this phenomenon would not, however, reduce
the original estimates of Cl^{36} concentrations in thermally graphites, such as
those used exclusively in B, D, and F Reactors.

The Cl^{36} inventories in DR, H, C, KW, and KE have been recalculated allowing
for the reduction in Cl^{36} concentration in the halogen purified graphites;
the revised estimates of Cl^{36} inventories are as follows:

Reactor	:	B	D	F	DR	H	C	KW	KE
Cl^{36}, Curies	:	42	34	33	26	17	12	52	54

Consideration of the environment in which the measured samples were exposed
during reactor operation indicates that the revised estimates are probably con-
servatively high.

Introduction

The previous estimate[a] of Cl^{36} activity in the graphite of the old Hanford
reactors was based on the unsubstantiated assumption that all of the chlorine,
originally present in the graphite, remained in place. Even at that time, there
was sufficient evidence to suggest that the estimates were quite conservative;
however, there was not sufficient evidence to justify applying a specific re-
duction factor to bring the estimates closer to what might be reasonably exp-
ected.

_____

[a] Memo from W. C. Morgan to R. A. Winship, "Estimated Chlorine-36 Concentration
in the Graphite of the Old Hanford Reactors," dated February 24, 1986.

J-001 (6/84)

In order to obtain improved estimates of the actual Cl^{36} activities, samples of graphite which had been removed from process channel 1880 at KW Reactor were analyzed for Cl^{36}. Two procedures were used to determine Cl^{36} activities in the samples.[b] First, attenuation measurements were made to confirm the presence of a 0.7 MeV beta-emitter; this procedure yields a preliminary estimate of Cl^{36} activity, but the estimate would be too high if other sources of beta-rays of about 0.7 MeV were present. The more precise technique involved activating the residual Cl^{37} to Cl^{38} for use as a tracer; all chlorine isotopes were then chemically separated from the graphite. The Cl^{36} activity of the separated chlorine was then measured, and corrected for the separation efficiency measured on Cl^{38}.

Results

The original estimate of Cl^{36} activity was based on the assumption that all the chlorine remained in the graphite. The resultant Cl^{36} activity at position x in KW Reactor can be calculated as follows:

$$A_x = \frac{[Cl^{35}] \, N_o \, \lambda}{M \cdot R} (1 - e^{-\sigma \phi x}) \qquad (1)$$

Where:
A_x = Cl^{36} activity, $\mu Ci/g$, at position x,

$[Cl^{35}]$ = concentration of Cl^{35} originally in the graphite, ppm,

N_o = Avogadro's number, 6.023×10^{23}/mole,

λ = decay constant of Cl^{36}, 7.09×10^{-14}/sec,

M = atomic weight of chlorine, 35.45 g/mole,

R = conversion factor from disintegration per second to Curies, 3.7×10^{10}/sec,

σ = activation cross section for the Cl^{35} (n,γ) Cl^{36} reaction, 43 barns, and

ϕ_x = thermal neutron fluence at position x.

All of the process tube blocks in the active zone of KW Reactor were made of TSGBF graphite (Fowler, 1959). Samples of TSGBF were not available when the measurements of chlorine content were made; however, grade SGBF contained 7.8 ppm chlorine. SGBF was produced, by Speer Carbon Co., from the same raw materials, and to the same specifications, as National Carbon Co. used to produce TSGBF; thus, impurity contents should be the same for these two grades. Therefore, we used 7.8 ppm chlorine (75.77% Cl^{35}) as the original concentration in the tube block graphite.

(b) All measurements were performed by C. W. Thomas, Analytical Chemistry Section, Chemical Sciences Dept., PNL.

Morgan and Bunch (1960) determined that the thermal neutron flux along a tube channel, as a function of distance from the center line of KW Reactor, can be described by the following equation:

$$\phi_x = \phi_c \cos (0.484x)° \tag{2}$$

Where: ϕ_x = thermal neutron flux at position x,

ϕ_c = thermal neutron flux at the center of KW Reactor, and

x = distance (inches) from the center line.

The fluence at position x, Φ_x, is merely $\phi_x t$, where t is the time, sec, that the reactor operated; thus, we can write an equation for Φ_x in analogy with eq. 2:

$$\Phi_x = \Phi_c \cos (0.484x)° \tag{3}$$

Where: Φ_c is the thermal neutron fluence at the center of KW Reactor, $5.23 \times 10^{22}/cm^2$.

Equation 3 can be used to calculate Φ_x at the positions where the graphite samples were obtained and, then, the estimated Cl^{36} activity can be calculated using eq. 1. Table 1 compares the measured and calculated activities, using this procedure.

TABLE 1. Comparison of Measured and Calculated Cl^{36} Activities

Sample Number	Distance from Centerline, in.	Fluence $10^{22}/cm^2$	Sample Weight, g	Cl^{36} Activity, $\mu Ci/g$ Calc., A_c	Cl^{36} Activity, $\mu Ci/g$ Meas., A_m	Ratio A_c/A_m
G-1	79	4.11	1.342	0.160	0.0143	11.2
G-2	87	3.88	0.0674	0.156	0.0129	12.1
G-3	103	3.37	0.0055	0.147	0.0075	19.6
G-4	116	2.91	0.0624	0.137	0.0096	14.3
G-5	139	2.02	0.0283	0.112	0.0137	8.2
G-6	159	1.18	0.0059	0.077	0.0086	9.0

One might expect the ratio A_c/A_m to vary as a function of neutron fluence; for example, if neutron collisions with the atoms broke Cl-C bonds, the chlorine might then be free to diffuse out of the graphite and be swept out of the radiation zone. If all isotopes of chlorine have an equal probability of escaping, then the change in concentration of chlorine would be described by the differential equation:

$$\frac{dC}{dt} = CK\phi \tag{4}$$

Where: dC/dt is the rate of change in the total chlorine content,
C is the concentration at time t,
K is a constant, and
ϕ is the neutron flux intensity.

The solution for eq. 4, evaluated at $t = 0$ and $t = \infty$, is:

$$C = C_0 \, e^{-k\Phi}, \text{ or}$$

$$C_0/C = e^{k\Phi} \tag{5}$$

Where: C_0 = the chlorine content at t=0, and
$\Phi = \phi t$, the neutron fluence.

If we accept the premise that Cl^{35} and Cl^{36} have equal probability of escaping, then it follows that $A_c/A_m = C_0/C$, and eq. 5 can be written as:

$$A_c/A_m = e^{k\Phi} \tag{6}$$

Evaluating eq. 6 at $\Phi = 4.11 \times 10^{22}/cm^2$ and $A_c/A_m = 11.2$, we obtain $k = 0.5878 \times 10^{-22} cm^2$, using this value for k, we calculate the ratios A_c/A_m for samples G-2 to G-6, incl., as: 9.8, 7.2, 5.5, 3.3, and 2.0 respectively. Clearly, the ratios A_c/A_m in Table 1 do not show this dependence on fluence and, in fact, appear to be independent of fluence.

We will return to this question of dependence on fluence in the next section; for the moment, however, we will treat the ratio A_s/A_m as a constant for all six samples.

The impurities in graphite tend to cluster; thus, the smaller the sample, the less likely that it will be representative of the bar from which it was taken. It is, therefore, not surprising that the largest departure of the ratio A_c/A_m from the average ratio (12.4) occurs for the three smallest samples. Because of this increased variability with decreased sample size, a weighted average was calculated for samples G-2 to G-6, inclusive, as follows:

$$\Sigma A_c W/\Sigma A_m W = 23.4956 \times 10^{-3}/1.9482 \times 10^{-3} = 12.1$$

Where: A_c and A_m are the calculated and measured activities, respectively, and W is the sample weight.

A simple average of this weighted average ratio and the ratio for sample G-1 yields a value of 11.6 ± 0.6 for the best estimate of the ratio of calculated activity/measured activity for these samples.

Discussion

The apparent constancy of the ratio A_c/A_m for TSGBF would be expected if the remaining chlorine is trapped in closed porosity within the graphite.

Let us assume that the chlorine was initially chemically bound to carbon atoms at the walls of the interconnected porosity in the graphite. This assumption seems reasonable in view of the low ash content of the purified graphites; in fact, the chlorine contents of CSF and KCF greatly exceeded their ash concentrations. It is also consistent with the accepted view that the chlorine is a residual from the gas purification process, and is not easily desorbed.

During operation of the reactor, the radiation field disrupts these chemical bonds and allows the chlorine to migrate out of the graphite (as Cl_2, Cl-C compounds, etc.); however, at the same time the neutron radiation causes the graphite crystallites to expand. In some places, expansion of the crystallites closes off sections of porosity, thereby trapping part of the chlorine in a closed-pore (see, for example: Spalaris, Bupp, and Gilbert, 1957).

Early in the operating life of a reactor, all of the chlorine in the open pores will have diffused out of the graphite, leaving some fraction trapped in the closed pores. If most of the original chlorine was contained in sections of the pores having small openings connecting them to other pores, as seems likely, then the fraction of the chlorine trapped would be larger than the fraction of pore volume closed off during irradiation. If, on the other hand, the chlorine was uniformly distributed on the walls of the pores, the fraction of the chlorine trapped should be proportional to the initial reduction in surface area. Spalaris, Bupp, and Gilbert (1957) report data which indicate a 27 percent reduction in surface area[c] of a KC graphite sample after only 0.8×10^{20}/cm² total neutron fluence,[c] and that about 12 percent of the open porosity of a CSF sample was converted to closed porosity after 4.8×10^{20}/cm² total fluence.[d]

Spalaris, Bupp, and Gilbert (1957) also report that the fractional change in surface area is a linear function of fluence from about 0.5 to 7×10^{20}/cm²; their data indicate, however, that there must be a more rapid decrease in surface area at lower fluences. Moreover, they report that the fractional decrease in surface area at any given fluence is independent of the graphite grade; thus, if the chlorine is uniformly distributed on the pore walls, the fraction trapped should also be independent of graphite grade.

Note, however, that the chlorine in thermally purified graphites is due to residual impurities which were present in the starting materials; thus, they are probably dispersed in the impurity agglomerates, and probably do not have direct access to the open pore system. The reduction factor of 11.6, which should apply to all halogen purified graphites, will not apply to the chlorine in the thermally purified graphites.

One other fact is worth noting; that is: process channel 1880 at KW Reactor was an untubed channel, used as an oxidation monitoring facility; therefore, it sustained a higher thermal neutron fluence than did any of the other graphite in KW Reactor. Because it is the thermal neutrons that initiate the Cl^{35} (n,γ) Cl^{36} reaction, the higher neutron fluence implies a greater concentration of Cl^{36} than would be present in the remainder of the graphite. No allowance has been made for the increased thermal fluence sustained by the samples from process channel 1880, or for the decreased thermal fluence sustained by graphite near the fueled process tubes. Therefore, the true ratio of A_c/A_m, averaged over all of the graphite, may be somewhat higher than 11.6.

(c) Thermal neutrons are the major component of the total fluence.
(d) These irradiations were performed at low temperatures; higher fluences would probably be required to produce the same fractional changes at reactor operating temperatures.

Revised Estimate of Cl^{36} Inventory

Only thermally purified graphites were used in B, D, and F Reactors; therefore, we have no reason to modify the original estimate of Cl^{36} inventory in those three reactors. Tables 2 and 3 show the graphite grades allocated to the various zones in the remaining five reactors; it can be seen that halogen purified (grade designations ending with "F") graphites were used to varying degrees in the different zones of these reactors.

TABLE 2. Approximate Graphite Allocation in DR, H, and C Reactors

	Graphite Grades[1]		
Zone	DR Reactor	H Reactor	C Reactor
Green	86% KCF, 13% CSF, 1% KSF	68% CSF, 17% GBF, 15% KCF	GBF
White	40% KC, 20% KS, 14% CS, 15% CSF, 11% KCF	87% CSF, 12% GBF, 1% KCF	GBF
Blue	66% CS, 22% KC, 12% KS	87% CS, 13% GBF	GBF
Red	71% CS, 17% KC, 12% KS	48% CS, 45% KS, 7% Misc.[2]	93% CS, 7% KS

(1) Grade designations which indicate position in a heat (such as KCNF, CSNF, CSO, etc.) are tabulated as part of the primary grade (KCF, CSF, CS, etc.).
(2) Listed as "Group II," no other identification; these may be early F processed bars with high-chlorine content.

TABLE 3. Graphite Allocation for KW and KE Reactors

		Graphite Grades	
Zone	% of Stack	KW	KE
Green	40	68% TSGBF, 32% CSGBF[1]	57% TSGBF, 43% SGBF
Blue	30.5	63% TSGBF, 37% CSGBF[2]	TSGBF
Red	29.5	55% TS-AGOT, 37% CS, 5% KC[3]	85% TS-AGOT, 15% TSGBF[4]

(1) Includes 10% CHF; CHF is a CS graphite, extruded at Clarksburg in 1944 and CBF processed in 1948.
(2) Includes about 1% CHF and 0.5% CSF.
(3) Remaining 3% is CSGBF plus a few bars of TSGBF and KCF.
(4) Includes a few bars of CSGBF.

Applying a reduction factor of 11.6 to the original chlorine content of the halogen purified graphites, and summing the fractional contributions of all graphite grades used in a reactor zone, we obtain the "effective" concentrations of Cl^{35}, shown in Table 4. By effective, we mean the amount which is assumed to either be associated with impurity agglomerations, or trapped in closed pores after an initial irradiation period.

TABLE 4. Effective Concentrations of Chlorine-35 by Reactor Zone

	Chlorine-35 Concentration, ppm			
Reactor	Green	White	Blue	Red
B	2.96	2.84	1.67	2.96
D	2.96	1.67	1.67	2.96
F	2.58	2.49	1.67	2.96
DR	0.54[1]	2.25[1]	2.10	2.04[2]
H	0.59[1]	0.63[1]	1.51	2.22[2]
C	(3)	0.46	0.46	1.76[4]
KW	(3)	(3)	0.49	2.55[4]
KE	(3)	(3)	0.50	2.59[4]

(1) The dih of the CSF bars was too high to justify use of 20 ppm chlorine, therefore, the CSF in these zones was assumed to be 10 ppm.
(2) "Group-II" was assumed to have high chlorine content; assumed 20 ppm Cl as an average.
(3) Green and White zones were combined for C Reactor; Green, White, and Blue were combined for KW and KE.
(4) Chlorine content of TS-AGOT assumed the same as in KSO, 3.9 ppm.

The equations and various factors required to calculate Cl^{36} concentrations and inventories are given in the memo to Winship,[a] and are not reproduced here. Tables 5 and 6 show the resultant Cl^{36} concentrations and inventories by reactor zone.

TABLE 5. Chlorine-36 Concentration by Reactor Zone·

	Chlorine-36 Concentration, ppm			
Reactor	Green	White	Blue	Red
B	1.61	1.33	0.56	0.10
D	1.57	0.76	0.54	0.09
F	1.22	1.01	0.47	0.08
DR	0.23	0.81	0.53	0.05
H	0.27	0.25	0.52	0.23
C	--	0.23	0.19	0.23
KW	--	--	0.37	1.25
KE	--	--	0.39	1.32

TABLE 6. Inventory of Chlorine-36 by Reactor Zone

	Chlorine-36 Activity, Curies				
Reactor	Green	White	Blue	Red	Total
B	10.5	17.5	13.5	1.0	42.4
D	10.2	9.9	12.9	0.9	33.9
F	8.0	13.3	11.2	0.8	33.3
DR	1.6	10.7	12.7	0.5	25.5
H	1.4	2.8	8.8	4.3	17.3
C	--	3.7	3.5	4.4	11.6
KW	--	--	21.1	31.2	52.3
KE	--	--	22.4	31.9	54.3

REFERENCES

Morgan, W. C., and W. L. Bunch. 1960. Neutron Flux as a Function of Tube Power, HW-63991

Spalaris, C. N., L. P. Bupp, and E. R. Gilbert. 1957. "Surface Properties of Irradiated Graphite," J Phys Chem, 61:350.
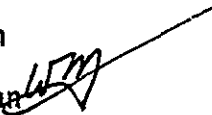
APPENDIX D

SOURCE OF TRITIUM IN GRAPHITE

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Battelle
Pacific Northwest Laboratories

Date   October 3, 1985

To   M. L. Smith

From   W. C. Morgan

Subject   Extrapolation of DR Reactor Core
Activities to Other Reactors

Although the graphites used in the oldest reactors were produced by the same manufacturing process, impurity contents of these graphites can vary significantly both within a grade of graphite and between grades. Very little data is available on elemental composition of the impurities; however, some appreciation of the differences can be gained from the changes in nuclear purity. Nuclear purity, measured in delta-in-hours (dih), is most heavily influenced by the concentrations of boron, the rare earths, vanadium, and titanium; however, for the same starting materials and purification process, total impurity content seems to have been well correlated with dih.

In the older piles, graphite was sorted by dih; that with the highest dih (highest purity) was used in the central "Green Zone," slightly less pure material was used in the "White Zone," even less pure in the "Blue Zone," and the remainder was relegated to the outer "Red Zone." For B Reactor, graphite in the Green Zone averaged +0.255 dih; in D, 75% averaged +0.309 dih (the remainder averaged +0.203 dih); and in F, 29% averaged +0.334 dih, with the remainder measuring +0.309 dih (however, a "new batch" of KS became available with a dih of +0.622, and some of this was used in the White Zone).

In contrast, graphite in the Green Zone at DR averaged +0.946 dih, and graphite in the Red Zone averaged only slightly lower dih (-0.30 vs. -0.25) than some of that used in the White Zone at B Reactor. More important, all of the graphite used in the Green Zone, and some of that in the White Zone at DR was F-Processed; the F purification process reduced impurity levels by an order of magnitude, or more, even on the early batches.

Variability in source of the cokes, hence, variability in impurity concentrations, introduces another potential problem in extrapolating measurements at DR to the other reactors. Part of this uncertainty could be removed by elemental analysis of the graphites used to construct the different reactors. I have archived one or more bars of most of the graphite grades used in the Hanford reactors; sections of these bars could be analyzed to determine relative concentrations of the important elements.

I should also mention that the tritium in the graphite probably is primarily a result of the reaction of fast neutrons with nitrogen: $_1N^{14} + \eta \rightarrow C^{12} + H^3$, which is the source of H^3 in our atmosphere. Thus, like C^{14}, its distribution in the graphite may be dependent on impurity concentrations, temperature gradients which existed during plant operation, etc.; however, because of the difference in the energies of the neutrons required for the reactions, the ratio of C^{14} to H^3 should vary with proximity to the fuel.

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APPENDIX E

RATIO OF ^{59}Ni TO ^{63}Ni

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Ratio of ^{59}Ni to ^{63}Ni

The ratio of ^{59}Ni to ^{63}Ni may be represented by the expression:

$$\frac{^{59}Ni}{^{63}Ni} = \frac{\lambda 59}{\lambda 63} \cdot \frac{\sigma 58}{\sigma 62} \cdot \frac{58}{62} \quad \text{Equation A}$$

where:

λ_{59} is the mean life of ^{59}Ni $= \dfrac{.693}{7.6 \times 10^4}$ yr^{-1}

λ_{63} is the mean life of ^{63}Ni $= \dfrac{.693}{100}$ yr^{-1}

$\dfrac{N_{58}}{N_{62}}$ is the ratio of target isotopes of Nickel $= \dfrac{68.27}{3.59}$

σ_{58} is thermal absorption cross-section of ^{58}Ni $= 4.6$ b

σ_{62} is thermal absorption cross-section of ^{62}Ni $= 14.5$ b

Substituting into Equation A

$$\frac{^{59}Ni}{^{63}Ni} = \frac{9.12 \times 10^{-6}}{6.93 \times 10^{-3}} \cdot \frac{4.6}{14.5} \cdot \frac{68.27}{3.59} = 0.0079$$

E-1

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APPENDIX F

CONTAMINATED FACILITY OUTSIDE THE REACTOR BLOCK
AND FUEL STORAGE BASINS

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ESTIMATED RADIONUCLIDE INVENTORY OF GROUND
DISPOSAL FACILITIES UNDER A MOUND OVER
A REACTOR ASSUMING A 3 TO 1 SLOPE

The in situ option using a mound with a radius of 210 feet and a 3 to 1 slope
is assumed. The ground disposal facilities under the mound are as follows:
116-B-3 Pluto Crib; 116-B-4 dummy decontamination tank drain; 116-D-6 105-D
Cushion Corridor French Drain; 116-F-11 105-F Cushion Corridor French Drain;
116-KE-1 115 Crib; 116-KE-3 105-KE Storage Basin Drain; 116-KW-1 115 Crib;
116-KW-2 105-KW Storage Basin Drain; and 105-C Ball 3X disposal silos.

The curie burdens estimated for these facilities are listed below:

Facility Identification	Radionuclides	Estimated Curies* (March 1985)
116-B-3 Pluto Crib	^{3}H	0.22
	^{90}Sr	0.02
	^{137}Cs	0.14
	^{152}Eu	0.26
	^{154}Eu	0.03
	^{238}U	0.0003
	^{238}Pu	0.0001
	^{239}Pu	0.0006
116-B-4 Dummy Decon Drain	^{60}Co	0.02
116-D-6 105-D Cushion Corridor French Drain	Mixed	<1
116-F-11 105-F Cushion Corridor French Drain	Mixed	<1
116-KE-1 Crib	^{3}H	80
	^{14}C	110
	Others	1
116-KE-3 105-KE Storage Basin Drain	Mixed	<1
116-KW-1 Crib	^{3}H	80
	^{14}C	110
	Others	1
116-KW-2 105-KW Storage Basin Drain	Mixed	<1
105-C Ball 3X Silos**	^{60}Co	80
	^{63}Ni	2

*Inventories based on data in UNI-946 and decayed to March 1, 1985.

**This is the only ground disposal facility that may be disturbed should
the one-piece reactor dismantlement alternative be selected.

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APPENDIX G

HAZARDOUS MATERIAL INVENTORY

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DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE		COMMENTS
				YES	NO	
9/25/85	Front far side 0'	Lead	700 lbs	x		Lead brick
9/25/85	Front far side -9'	Lead-Cadmium	500 lbs	x*		Monitoring tube
9/25/85	Front far side -9'	Lead	7,000 lbs	x		Cave (lead brick)
9/25/85	Front far side -9'	Lead	1,000 lbs	x		Lead brick
9/25/85	Front far side X-1	Lead	500 lbs	x*		Shield plugs
9/25/85	Top of Reactor	Lead	3,500 lbs	x		7 rod tip shields
9/25/85	Rod Room - outer	Lead	625 lbs	x		Lead brick
9/25/85	Rod Room - inner	Lead	1,000 lbs	x		Lead brick
9/25/85	Sample rooms	Lead	2,000 lbs	x		Lead shot in sample trays
9/25/85	Rear face tool room	Lead	50 lbs	x		Lead sheet
9/25/85	Rear face +10'	Lead	5 lbs	x		Lead wool
9/25/85	Elevator Eqpt Level	Lead	1 ea	x		Mercury switch
9/25/85	Inside Reactor Block	Lead	160,000 lbs		x	

FACILITY 105-C

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	NO	COMMENTS
9/26/85	Front far side X-1	Lead	1,750 lbs	x		Lead brick
9/26/85	Front far side X-2	Lead	500 lbs	x*		Shielded door
9/26/85	Front far side X-2	Lead	3,500 lbs	x		Lead brick
9/26/85	Top of Reactor	Lead	9,100 lbs	x*		Cave (lead brick)
9/26/85	Top of Reactor	Lead	4,200 lbs	x*		Ball cave
9/26/85	Top of Reactor	Lead	4,200 lbs	x*		Rod tip shield
9/26/85	Rear of Reactor	Lead	26,000 lbs	x*		Shielding Door
9/26/85	Rear of Reactor	Lead	800 lbs	x*		Central Viewer
9/26/85	Inside Reactor Block	Lead	160,000 lbs		x	

9 2 1 2 5 0 9 3 7

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	REMOVABLE NO	COMMENTS
10/1/85	Rod room - inner	Lead	625 lbs	x		Lead brick
10/1/85	Rod room - outer	Lead	50 lbs	x		Lead brick
10/1/85	Rod room - outer	Lead	500 lbs	x		2 shield plugs
10/1/85	Front far X-2	Lead	200 lbs	x		Lead brick
10/1/85	Top of Reactor	Lead	4,900 lbs	x		Rod tip shields
10/1/85	Top of Reactor	Lead	625 lbs	x		Loose lead brick
10/1/85	Top of Reactor	Lead	10,000 lbs	x		Cave (lead brick)
10/1/85	Top of Reactor	Lead	12,000 lbs	x		Ball cave (lead brick)
10/1/85	Front far -9'	unknown	?	x		Two tanks - approx. 3" of liquid in one tank. Contents of second tank unknown.
10/1/85	Inside Reactor Block	Lead	160,000 lbs		x	

FACILITY __105-DR__

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	REMOVABLE NO	COMMENTS
10/1/85	Sample rooms	Lead	3,500 lbs	x		Lead shot in sample trays
10/1/85	Rod room - inner	Lead	500 lbs	x		Lead brick and shield plugs
10/1/85	Rod room - outer	Lead	550 lbs	x		2 shield plugs
10/1/85	Top of Reactor	Lead	10,500 lbs	x		2 caves (lead brick)
10/1/85	Top of Reactor	Lead	8,400 lbs	x		Ball cave (lead brick)
10/1/85	Top of Reactor	Lead	3,000 lbs	x		Rod tip shields & miscellaneous lead brick and sheet
10/1/85	Front far X-1	Lead	700 lbs	x		Lead brick and large pig
10/1/85	Front far X-2	Lead	150 lbs	x		Lead brick and sheet
10/1/85	Front far X-3	Lead	1,000 lbs	x		Lead brick and sheet
10/1/85	Inside Reactor Block	Lead	160,000 lbs		x	

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	NO	COMMENTS
10/8/85	Far side -9'	Lead	60 lbs	x		Lead plate
10/8/85	Far side X-1	Lead	1,400 lbs	x		Lead brick shielded box
10/8/85	Far side X-1	Lead	60 lbs	x		Lead sheet on pipe
10/8/85	Far side X-1	Lead	60 lbs	x		Lead plate
10/8/85	Far side X-2	Lead	1,000 lbs	x		Lead brick - scattered
10/8/85	Far side X-2	Lead	120 lbs	x		Lead sheet on pipe & lead plate
10/8/85	Far side X-2	Lead	20 lbs	x		Lead wool
10/8/85	Far side X-2	Lead	500 lbs	x		Lead brick around tube
10/8/85	Far side X-2	Cadmium	25 lbs	x		Neutron shield bonnets (2)
10/8/85	Far side X-2	Cadmium	5 lbs	x		Cadmium sheet
10/8/85	Top of Reactor	Lead	4,000 lbs	x		Rod tip shields (6)
10/8/85	Top of Reactor	Lead	4,000 lbs	x		Lead brick (airway to rear face)
10/8/85	Top of Reactor	Lead	9,000 lbs	x		Lead brick (2 caves)
10/8/85	Sample rooms	Lead	5,000 lbs	x		Lead shot in sample trays
10/8/85	Rod room - inner	Lead	125 lbs	x		Lead brick
10/8/85	Rod room - outer	Lead	2,100 lbs	x		Shields plugs
10/8/85	Near side -9'	Lead	300 lbs	x		Lead brick shielded box
10/8/85	Front face -0'	Lead	25 lbs	x		Lead brick on duct
10/8/85	Inside Reactor Block	Lead	160,000 lbs		x	

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UNI-3714 REV1

HAZARDOUS MATERIALS INVENTORY

FACILITY___105-H_____

*Removal requires
special equipment

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	NO	COMMENTS
10/8/85	Far side -9'	Lead	250 lbs	x		Lead brick & sheet
10/8/85	Far side X-2	Lead	200 lbs	x		Lead brick & sheet
10/8/85	Far side X-3	Lead	500 lbs	x		Lead brick
10/8/85	Far side X-3	Cadmium	20 lbs	x		Sheet around experiment
10/8/85	Top of Reactor	Lead	4,200 lbs	x		Rod tip shields
10/8/85	Top of Reactor	Lead	2,500 lbs	x		Lead brick
10/8/85	Top of Reactor	Lead	4,000 lbs	x*		Cave
10/8/85	Top of Reactor	Lead	4,000 lbs	x*		Ball cave
10/8/85	Far side	Lead	40,000 lbs	x*		Shield door (equip. hatch)
10/8/85	Sample rooms	Lead	4,000 lbs	x		Lead shot in sample trays
10/8/85	Near side -12'	Lead	750 lbs	x		Lead brick
10/8/85	Near side -12'	Lead	1,000 lbs	x		Lead sheet around piping
10/8/85	Rod room - inner	Lead	800 lbs	x		Lead brick & lead sheet
10/8/85	Rod room - outer	Lead	2,100 lbs	x		Shield plugs & shield
10/8/85	Near side stairway	Mercury	--	x		2 Mercury switches
10/8/85	Near side -12'	Mercury	--	x		3 Mercury switches
10/8/85	Near side -12'	Solvent	--	x		Tank (contents unknown)
10/8/85	Inside Reactor Block	Lead	160,000 lbs		X	

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	REMOVABLE NO	COMMENTS
12/19/85	Irradiation Test Rm.	Lead	500 lbs.	X		Lead Brick
"	-9' Near Side	Lead	1200 lbs.	X		Lead Brick
"	Inner Rod Room	Lead	100 lbs.	X		Lead Brick
"	Inner Rod Room	Lead	4200 lbs.	X*		Rod Tip Shield
"	Outer Rod Room	Lead	500 lbs.	X		Lead Brick & Sheet
"	X-1 Level	Lead	5000 lbs.	X		Lead Brick
"	X-1 Level	Lead	10,000 lbs.	X		6 Large Shielded Pigs
"	X-2 Level	Lead	2000 lbs.	X		1 Lead Pig
"	X-2 Level	Lead	750 lbs.	X		Lead Brick
"	X-2 Level	Lead	14,000 lbs	X*		Neutron Beam Shutter
"	Top of Reactor	Lead	5,000 lbs.	X		3 Ball Storage Containers
"	Top of Reactor	Mercury	50 ea.	X		Mercury Switches
"	Rear of Reactor	Lead	48,000 lbs	X*		Shielding Doors
"	Rear of Reactor	Lead	2,700 lbs	X*		Central Viewer
"	Inside Reactor Block	Lead	240,000 lbs		X	
	Note: Far side rooms were not inspected. Area is boarded up to discourage entry.					

FACILITY___105-KW_____

*Removal requires
special equipment

DATE	LOCATION	MATERIAL	ESTIMATED AMOUNT	REMOVABLE YES	NO	COMMENTS
12/19/85	-9 Landing	Lead	25 lbs.	X		Lead Brick
"	Outer Rod Room	Lead	500 lbs.	X		Lead Brick
"	Outer Rod Room	Lead	100 lbs.	X		Lead Sheet
"	Inner Rod Room	Lead	2500 lbs.	X		Lead Brick
"	Inner Rod Room	Lead	10,000 lbs.	X*		Rod Tip Shield
"	X-1 Level	Lead	750 lbs.	X		Lead Brick
"	X-1 Level	Lead		X		6 Shielded Pigs
"	X-2 Level	Lead	300 lbs.	X		Lead Sleeve
"	X-2 Level	Lead	5000 lbs.	X		Lead Brick
"	Top of Reactor	Lead	250 lbs.	X		Lead Brick
"	Top of Reactor	Mercury	47 ea.	X		Mercury Switches
"	Rear of Reactor	Lead	48,000 lbs	X*		Shielding Door
"	Rear of Reactor	Lead	2,700 lbs	X*		Central Viewer
"	Inside Reactor Block	Lead	240,000 lbs		X	

Note: Far side rooms were not inspected. Area is boarded up to discourage entry.